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Frequency combs in the XUV by intra-laser high harmonic generation for ultra-precise measurements of the fine structure constant

Thomas Sudmeyer
UNIVERSITE DE NEUCHATEL

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Final Report

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14. ABSTRACT
The program met the goal producing ultrahigh precision spectroscopy exploiting a new XUV source that I will realize in the framework of an ERC staring grant. The high harmonics generation was placed directly inside the intra-cavity multi-kilowatt beam of a femtosecond laser that operates at multi-megahertz repetition rates. While the ERC project has targeting the proof-of-principle demonstration of high power intra-laser HHG, with this proposal, we want to enable new directions in XUV frequency comb spectroscopy. In the first year, we will focus on understanding the optimum stability parameters of a thin disk laser in intracavity operation. We will experimentally investigate noise properties and target the demonstration of stable IR driving frequency comb at unprecedented high intracavity power levels.

15. SUBJECT TERMS
high resolution XUV-microscopy, Coherent vacuum- and extreme ultraviolet, frequency comb spectroscopy, femtosecond laser-driven high-harmonic, fine structure constant measurement, high harmonic generation HHG, table-top femtosecond sources, EOARD

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FINAL Report

Intra-laser ultra-precise measurements of the fine structure constant 122127

FA8655-12-1-2127

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PI’s Qualifications

Prof. Thomas Südmeyer (UNINE) studied physics at the Leibniz University Hanover and the Ecole Normale Supérieure (ENS) Paris. In 1999, during a six month EU fellowship at the Institute of Photonics, Strathclyde University in Glasgow, he started working in the area of ultrafast lasers. In 2003, he obtained the PhD degree from ETH Zurich for research on the first mode locked thin-disk lasers and the realization of novel nonlinear systems. From 2003 to 2005, he developed industrial laser solutions such as high-power 266-nm lasers in the Department of Photonics Research, Sony Corporation, Tokyo, Japan. From 2005-2011, he investigated new laser concepts for ultrafast science and technology, such as high power femtosecond diode-pumped solid-state lasers, multi-gigahertz repetition rate sources, and novel mode locked semiconductor lasers at ETH Zurich, obtaining the Habilitation degree in August 2011. Since September 2011, he is full professor and director of the Time and Frequency Laboratory at the University of Neuchatel, where he investigate novel approaches for frequency combs and their applications in metrology. His team consists of >20 researchers who have a strong accumulated experience in various areas of metrology, frequency comb development and evaluation, and laser molecular spectroscopy, both for laser stabilization and for trace gas sensing. The expertise in laser gas sensing covers various spectroscopic techniques, such as wavelength-modulation spectroscopy, photoacoustic spectroscopy, and quartz-enhanced photoacoustic spectroscopy, using various near- and mid-IR lasers sources, including QCLs. Furthermore, the UNINE group contributes to ESA and EU projects devoted to the development of future space optical clocks, such as EU FP7 project "SOC-2" and ESA-GSTP project "Optical Atomic Clock in Space". Thomas Südmeyer is the author of more than 50 papers in international peer-reviewed journals, two book chapters, and he holds or applied for 7 patents. He has been the Coordinator of the FP6 European Project MULTIWAVE and recently received an ERC Starting grant. The significance of his contributions to the optical community has been recognized by several elections to program committees, as program chair of the Europhoton 2012, conference chair of the Ultrafast Optics 2013, and as associate editor for Optics Express, which is the 3rd-highest ranked journal in Optics (ISI 2009).

University’s Qualifications

The Neuchatel region has an enormous knowledge base in metrology, microtechnology, and space applications. A healthy industry has developed thanks the expertise and innovative research, which enable to actively exploit new technology platforms. Several spin-offs of the University of Neuchatel are leaders in the global market. For example, all atomic clocks for the European satellite-based global positioning system GALLILEO were manufactured in Neuchatel. With the integration of the research activities of the former Neuchatel Observatory, the University of Neuchatel is at the center of this outstanding area of excellence. The University of Neuchatel provides outstanding research at the forefront of metrology, which is honored by numerous top level funding schemes, including ERC, FP7, ESA, and Nanotera.ch projects. It is extremely well embedded in the national and international research landscape, having collaborations with all major research institutions. Moreover, it has long-standing collaborations with all Swiss industrial key players and the support of Swiss administration offices like the Swiss space office, the METAS metrology institute and network agencies.

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Relevance to DOD missions

Increasing the precision and reducing the complexity of time and frequency measurements is at the core of numerous technologies with high relevance for DOD missions. The targeted development of a new tool for XUV comb generation and spectroscopy is expected to show a high synergy for the realization of numerous other exciting devices in various areas like advancement of sensors and nano-technological devices.

Summary of Project Succeses

Coherent vacuum- and extreme ultraviolet (VUV/XUV) light sources open up new opportunities for science and technology. The most promising technique for table-top sources is femtosecond laser-driven high-harmonic generation (HHG) in gases. Unfortunately, their VUV/XUV photon flux is not sufficient for most applications. This is caused by the low average power of the driving lasers and the extremely poor conversion efficiency. Furthermore, their low kilohertz repetition rate is not suitable for direct frequency comb spectroscopy, because the frequency comb spacing equals the repetition rate.

The program met the goal producing ultrahigh precision spectroscopy exploiting a new XUV source that I will realize in the framework of an ERC staring grant. The high harmonics generation is placed directly inside the intra-cavity multi-kilowatt beam of a femtosecond laser that operates at multi-megahertz repetition rates. While the ERC project is targeting the proof-of-principle demonstration of high power intra-laser HHG, with this proposal, we want to enable new directions in XUV frequency comb spectroscopy. In the first year, we will focus on understanding the optimum stability

parameters of a thin disk laser in intracavity operation. We will experimentally investigate noise properties and target the demonstration of stable IR driving frequency comb at unprecedented high intracavity power levels.

The final target was the precise measurement of the 1S-2S transition of He+, which has 40.8 eV energy, corresponding to 60.8 nm wavelengths for a two-photon transition. Precise spectroscopy of the 1s-2s transition for atomic hydrogen has been one of the most valuable tests for quantum electrodynamics (QED). He+ spectroscopy appears even more promising, as higher order perturbation terms for the self-energy correction of the electron and the vacuum polarization increase with the nuclear charge. Furthermore, as a charged particle, He+ can be easily captured in an ion trap and can be sympathetically cooled to reduce higher order Doppler shifts. In the first year, we will focus on understanding the optimum stability parameters of a thin disk laser in intracavity operation. We will experimentally investigate noise properties and target the demonstration of stable IR driving frequency comb at unprecedented high intracavity power levels.

Title of Proposal

Frequency combs in the XUV by intra-laser high harmonic generation for ultra-precise measurements of the fine structure constant

Requested Start Date

July 1 2012

Duration of Proposed Effort (in months)

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36 months

Abstract of proposed research effort

Coherent extreme ultraviolet (XUV) light sources open up new opportunities for science and technology. Promising examples are attosecond metrology, spectroscopic and structural analysis of matter on a nanometer scale, high resolution XUV-microscopy and lithography. Large-scale accelerator based sources have very limited accessibility, effectively preventing their use for the majority of research projects. The most promising technique for table-top sources is femtosecond laser-driven high-harmonic generation (HHG) in gases. However, their current XUV photon flux is not sufficient for most applications. This is caused by the low average power of the kHz repetition rate driving lasers (<10 W) and the extremely low conversion efficiency ($<10^{-6}$).

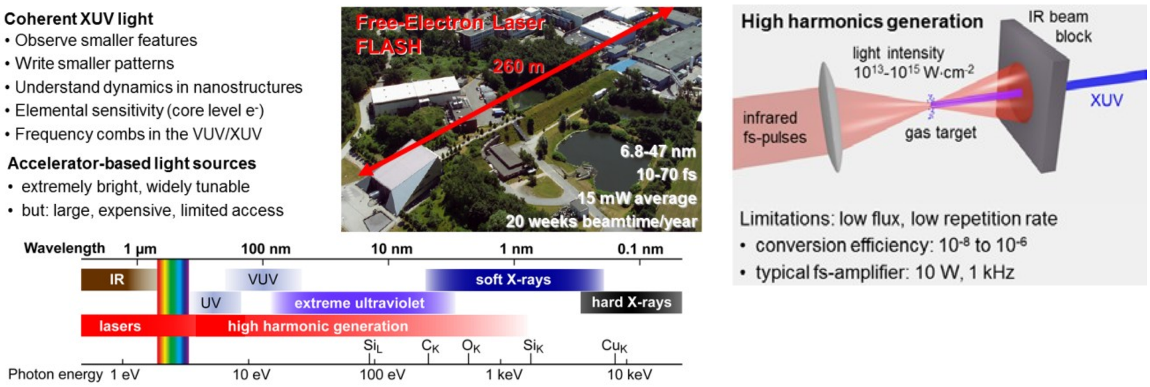

Fig. 1: Accessible wavelength range of laser-driven high harmonic generation

Unfortunately, up to now there are no simple solutions to substantially increase the efficiency of high harmonic generation. The low efficiency is cause by the phase-mismatch between generated XUV and the fundamental light, which in free space can only be slightly optimized by focus size and gas pressure.

The most successful method to increase the efficiency is the use of passive enhancement cavities, which allows increasing the conversion efficiency by increasing the field strength of the driving laser. Several research groups have demonstrated HHG at MHz repetition rates with systems based on Ti:Sapphire lasers or fiber lasers [1-5]. These systems typically used enhancement factors of several hundreds to thousands, which enables strong field physics already at moderate driving pulse energies. However, the enhancement cavity losses need to be minimized to the few per mille level for reaching sufficiently high finesse, which is difficult for kW intracavity power levels. Moreover, the input mode-matching is challenging and issues such as nonlinearities by the nonlinear interaction with the gas target, dispersion or transverse mode distortions at the high power levels are difficult to compensate for. In addition, the requirements on the phase stability of the driving laser systems are very demanding, even if complex state-of-the-art locking electronics is used.

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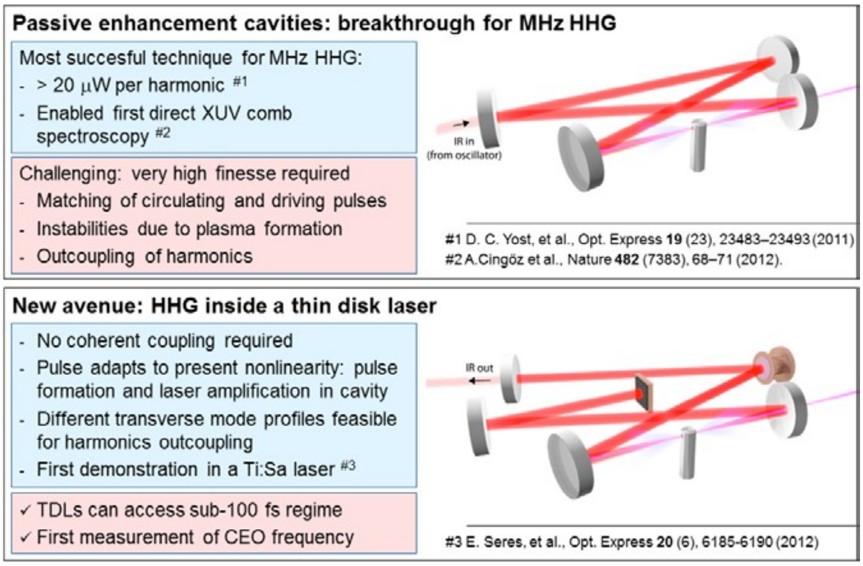

Fig. 2: Comparison between passive enhancement cavities and intra-laser HHG

In the project MEGA-XUV, for which I was awarded with an ERC starting grant in 2011, I target the realization of a simpler and more efficient source of high-flux XUV radiation. Instead of amplifying a laser beam to high power levels and dumping it after the HHG interaction, the generation of high harmonics is placed directly inside the intra-cavity multi-kilowatt beam of a femtosecond laser. Thus, the unconverted light is “recycled”, and the laser medium only needs to compensate for the low losses of the resonator. Achieving passive femtosecond pulse formation at these record-high power levels will require eliminating any destabilizing effects inside the resonator. In contrast to passive enhancement cavities [1-3, 6] where the circulating pulse has to match the driving pulses, both pulse formation and laser amplification are achieved inside the laser cavity, where the nonlinear process takes place. In addition, there is no need for coherent coupling of the driving pulses, which is a challenging point in passive enhancement cavities, and the circulating pulse can simply adapt to the present nonlinearity. Another potential advantage is that different transverse mode profiles can be achieved, for example TEM_{01}, for efficient output coupling of the high harmonics via a hole in a cavity mirror [7]. This is not the case in passive enhancement cavities, where efficient extraction of the UV radiation from these very high finesse cavities is challenging [8, 9].

A first proof-of-principle demonstration of intra-laser HHG demonstrated in 2012 by E. J. Seres et al. [10] using a Ti:Sapphire laser. However, the high nonlinearity of the laser material and the limited power levels are a strong challenge for further power scaling. Achieving self-starting femtosecond pulse formation at record-high average power levels in the kW regime will require the elimination of any destabilizing effects inside the resonator. This appears to be feasible only with the technology of ultrafast thin disk lasers, because all key components are used in reflection, as will be explained in the following.

In a typical modelocked oscillator, a single pulse propagates between the mirrors of an optical resonator. A part of its energy is transmitted through the output coupler, thus forming a pulse train at the output. The lost energy is recovered by the pulse amplification in the gain medium. The pulse formation is achieved by an intracavity saturable absorber. The most successful passive mode-locking technique for high power lasers is the semiconductor-saturable-absorber-mirror (SESAM, [11]). Driving a modelocked laser to high power levels is challenging due to thermal effects in the gain medium.

During my Ph.D., my colleagues and I developed a new class of power-scalable lasers, which since then achieved higher power levels and pulse energies than any other femtosecond oscillators. In 2000, we demonstrated the first SESAM modelocked thin disk laser (TDL) which increased the pulse energy

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of femtosecond diode-pumped solid-state lasers from below 10 nJ to 500 nJ [12] and the power from 1.4 W to 16 W.

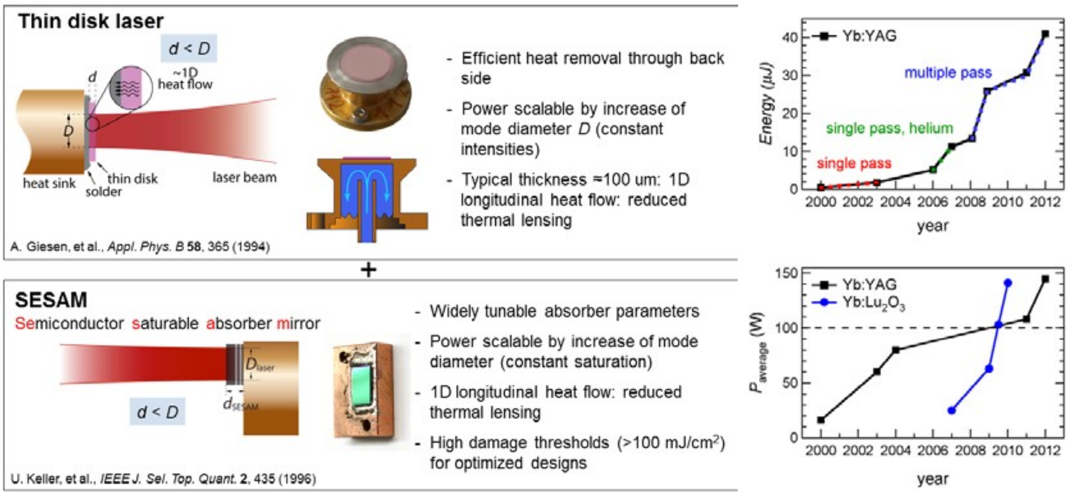

Fig. 3: SESAM-modelocked thin disk lasers achieve the highest pulse energies and average powers of any femtosecond laser oscillator.

The thin-disk geometry [13] solves the thermal problems occurring in conventional high-power rod or slab lasers. The gain medium has the shape of a very thin disk that is mounted directly onto a heat sink and used in reflection.

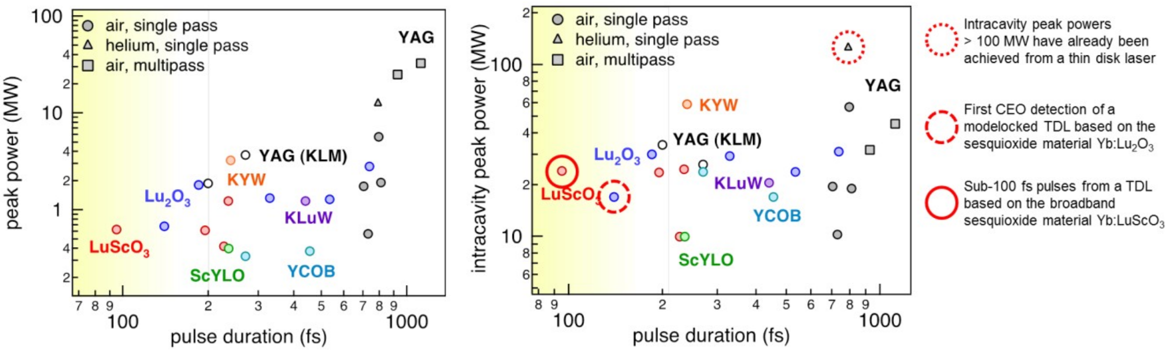

Fig. 4: Comparison of peak power and intra-laser peak power as function of pulse duration for ultrafast thin disk lasers. The circles in red indicate three major results, which illustrate the high potential of TDLs for intra-cavity HHG.

Today, pulse energies >40 µJ have been demonstrated and average powers of up to 145 W [14-16]. We achieved three major break-throughs, which indicate the high potential of intra-laser HHG (see figure 5):

- An Yb-YAG TDL operated in Helium already achieved a peak power >100 MW
- In 2011, we demonstrated the first CEO detection of at TDL
- Very recently, we achieved first TDL with sub-100 fs pulse duration

In my ERC project, I want to realize an oscillator with an intracavity average power above 5 kW at a pulse duration below 200 fs. The intracavity pulses will have an energy of >250 µJ and a peak power of >1 GW – a regime that has never before been achieved inside a modelocked resonator. Even at such power levels, the laser will support between 2% to 5% total intracavity losses, allowing for a large amount of freedom in the implementation of the HHG interaction and efficient XUV output coupling. The corresponding power loss of 100-250 W appears feasible in the thin disk laser technology.

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Assuming an intracavity focus with 30 µm diameter for the HHG interaction, the peak intensities would be >3·1014 W/cm², which is well-suited for HHG generation. The 1030-nm wavelength is longer than for the typically used 800-nm amplifier systems, leading to a higher cut-off energy [17], which at this intensity should be below a wavelength of 10 nm, respectively above photon energies of 100 eV. A conversion efficiency of 10^{-7} to 10^{-6} for the harmonics in the HHG plateau would correspond to average power levels between 0.5 – 5 mW per harmonic.

An extremely promising range of applications like VUV/XUV precision spectroscopy on He+ [18] or even exploring nuclear transitions [19] should in principle be feasible with such a table-top source. However, these important studies are not covered within the ERC grant, which targets the proof-of-principle demonstration of high power intra-laser HHG. With this proposal, we want to enable new directions in XUV frequency comb spectroscopy by exploiting the frequency comb properties of our new source. To this purpose, the laser has to be precisely CEP-stabilized, which should be feasible given our recent results (see figure 5). We will evaluate the noise properties in detail and optimize the laser parameter and stabilization schemes. In particular, we will evaluate the noise properties and stabilization limits in the presence of strong nonlinearities originating from the optical components and the ionized gas for HHG. In this way, we will realize a frequency comb in the XUV that is suitable for high precision XUV spectroscopy. The final target is the precise measurement of the 1S-2S transition of He+, which has 40.8 eV energy, corresponding to 60.8 nm wavelengths for a two-photon transition. Precise spectroscopy of the 1s-2s transition for atomic hydrogen has been one of the most valuable tests for quantum electrodynamics (QED). He+ spectroscopy appears even more promising, as higher order perturbation terms for the self-energy correction of the electron and the vacuum polarization increase with the nuclear charge. Furthermore, as a charged particle, He+ can be easily captured in an ion trap and can be sympathetically cooled to reduce higher order Doppler shifts. In the first year, we will focus on understanding the optimum stability parameters of a thin disk laser in intracavity operation. We will experimentally investigate noise properties and target the demonstration of stable IR driving frequency comb at unprecedented high intracavity power levels. In the second year, we will realize the HHG and characterize and optimized the XUV output for maximum radiation at 60.8 nm wavelength. In the third year, we will perform two-photon spectroscopy on He+.

Statement of Work (proposed services)

This project has a high synergy with my ERC starting grant MEGAXUV, in which I target the first demonstration of high-harmonic generation inside a high power ultrafast thin disk laser. The focus of this project is the exploitation of this work for the promising application of an ultra-precise measurement of the fine structure constant, which is not a research target of my ERC project.

As first step towards the realization of such measurement system, during the first year, we will investigate the feasibility of ultra-stable frequency comb generation in the infrared from a high power ultrafast thin disk laser. To this purpose, we will investigate the limiting factors and optimize its performance for high stability. In particular, we will investigate the influence of high nonlinearities inside the laser resonator, which can occur due to nonlinearities in dielectric mirrors and the high harmonic generation. In the second year, we will characterize and optimize the generated frequency comb in the VUV/XUV for the target application that requires radiation at 60.8 nm. To this purpose, we will use an XUV grating spectrometer. Particular challenging with be the precise calibration of the power levels in the individual harmonics, which will be independently verified with a calibrated Si photodiode. In the third year, we will optimize the system for precision spectroscopy on He+. To this purpose, we will use a vacuum system containing a Paul-trap for the He+ ions for high-precision two-photon spectroscopy.

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List of estimated expenses and costs in detail (US$)

Year 1 - totals

Item	cost per item in CHF	cost per item in USD	Total cost, USD	Notes
Expendable supplies and materials			**$ 15,640.00**	
Highly nonlinear photonic crystal fiber (from NKT Photonics, types SC-5.0-1040-PM, NL-1050-ZERO-2)	11,230.47	$11,500.00		for optimizing the supercontinuum generation necessary for CEP measurement and frequency comb stabilization
CCD camera for beam profile measurement (from Dataray, type Wincam D UCD15-1310)	4,042.97	$ 4,140.00		necessary for measuring the beam profile for the PCF fiber coupling
Equipment			**$ -**	
(none)				
Travel			**$ 8,000.00**	
Project result dissemination at major international conferences				2 postdoc visits, 1 visit project leader to international conferences (Photonics West, CLEO/US)
Publications and reports			**$ 3,486.00**	
2 open-access publications in Optics Express				Open access journal of the Optical Society of America
Total staff & student costs (direct + indirect)			**$ 22,874.00**	
Post-doc (= research assistant)	17,754.88	$18,181.00		2 person months for the CEP measurement and the optimization of SCG in a photonic crystal fiber for maximum frequency stability.
Experienced Technician	4,583.01	$ 4,693.00		0.6 person months for realization of a suitable mechanical setup and optimization of the frequency stabilization electronics.
Fee or profit			**$ -**	
Total cost			**$ 50,000.00**	

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Year 1 – Rate breakdown - staff cost

Cost per person month	Direct salary cost (incl. national insurance, etc.) or stipend (CHF)	Indirect cost (CHF)	Total cost per year (CHF)	Total cost per year (USD)	Percentage time (over 12 months)	Total staff cost (USD)
Post-doc (= research assistant)	88,776.35	17,755.27	106,531.62	$109,088.38	16.67%	$18,181.00
Experienced Technician	82,267.90	16,453.58	98,721.48	$101,090.80	4.64%	$ 4,693.00

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Year 2

Item	cost per item in CHF	cost per item in USD	Total cost, USD	Notes
Expendable supplies and materials			**$ 13,056.00**	
Calibrated VUV/XUV detector	8,000.00	$ 8,192.00		Precise calibration of the XUV flux.
Electron Multiplier Tube	4,750.00	$ 4,864.00		Measurement and optimization of the XUV spectrum.
Equipment			**$ -**	
(none)				
Travel			**$ 8,000.00**	
Project result dissemination at major international conferences				2 postdoc visits, 1 visit project leader to international conferences (Photonics West, CLEO/US)
Publications and reports			**$ 1,743.00**	
1 open-access publications in Optics Express				Open access journal of the Optical Society of America
Total staff & student costs (direct + indirect)			**$ 27,201.00**	
Post-doc (= research assistant)	10,110.35	$10,353.00		1.1 person months for optimization of the XUV spectrometer and precise characterization of the generated XUV output
Experienced Technician	16,453.13	$16,848.00		2 person months for optimizing the mechanical setup of the HHG chamber and the XUV spectrometer
Fee or profit			**$ -**	
Total cost			**$ 50,000.00**	

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Year 2 – Rate breakdown - staff cost

Cost per person month	Direct salary cost (incl. national insurance, etc.) or stipend (CHF)	Indirect cost (CHF)	Total cost per year (CHF)	Total cost per year (USD)	Percentage time (over 12 months)	Total staff cost (USD)
Post-doc (= research assistant)	91,165.10	18,233.02	109,398.12	$112,023.67	9.24%	$10,353.00
Experienced Technician	82,267.90	16,453.58	98,721.48	$101,090.80	16.67%	$16,848.00

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Year 3

Item	cost per item in CHF	cost per item in USD	Total cost, USD	Notes
Expendable supplies and materials			**$ 8,704.00**	
Mirrors for focusing of the XUV light	8,500.00	$ 8,704.00		Optical components for the high-precision XUV spectroscopy measurement
Equipment			**$ -**	
(none)				
Travel			**$ 8,000.00**	
Project result dissemination at major international conferences				2 postdoc visits, 1 visit project leader to international conferences (Photonics West, CLEO/US)
Publications and reports			**$ 1,743.00**	
1 open-access publications in Optics Express				Open access journal of the Optical Society of America
Total staff & student costs (direct + indirect)			**$ 31,553.00**	
Post-doc (= research assistant)	27,349.61	$28,006.00		3 person months for optimization of the XUV system for high coherence and flux at a wavelength of 60.8 nm. Realization of the measurements for the He+ spectroscopy.
Experienced Technician	3,463.87	$ 3,547.00		0.4 person months for mechanical connection of the intra-laser HHG vacuum system to the XUV precision spectroscopy setup
Fee or profit			**$ -**	
Total cost			**$ 50,000.00**	

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Year 3 – Rate breakdown - staff cost

Cost per person month	Direct salary cost (incl. national insurance, etc.) or stipend (CHF)	Indirect cost (CHF)	Total cost per year (CHF)	Total cost per year (USD)	Percentage time (over 12 months)	Total staff cost (USD)
Post-doc (= research assistant)	91,165.10	18,233.02	109,398.12	$112,023.67	25.00%	$28,006.00
Experienced Technician	82,267.90	16,453.58	98,721.48	$101,090.80	3.51%	$ 3,547.00

Total Cost of Proposal (US$)

$150,000

Payment Information

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Professional and academic experience

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6/1999 - 8/2003	PhD student, Physics Department, **ETH Zurich** (CH)
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9/2005 - 8/2011	Habilitation in physics, **ETH Zürich** (CH)
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Honors and awards

2011	**ERC Starting Grant of the European Union**
1998	**Leonardo fellowship of the European Union**
1995-1998	**Fellowship of the "Studienstiftung des deutschen Volkes"**

Professional services

Associate editor for Optics Express (2009-present)

Sub-Committee chair Europhoton (2012). Co-chair LPHYS (2009-2012).

Program committee Europhoton (2008-2010), ALT (2012), CLEO/Europe (2009-2011), ASSP (2012)

Publication record

2 book chapters, >50 publications in refereed journals, >1100 citations, >40 invited presentations

Hirsch H-index of 20

7 patents / patent applications

Research interests

Metrology: Realization and optimization of novel frequency comb technologies. Extension of frequency combs into new spectral regions (VUV/XUV, mid-IR). New approaches for frequency spectroscopy.

Ultrafast lasers: High power thin disk lasers (TDLs), gigahertz repetition rate diode-pumped solid-state lasers (DPSSLs), semiconductor disk lasers (SDLs), waveguide lasers.

Ultrafast nonlinear optics: Frequency conversion, pulse compression, ultrafast nonlinear dynamics. Generation of coherent radiation in the extreme ultraviolet (XUV) at multi-megahertz repetition rates.

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THOMAS SÜDMEYER CURRICULUM VITAE JANUARY 2012

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Frequency combs in the XUV by intra-laser high harmonic generation for ultra-precise measurements of the fine structure constant

Prof. Thomas Südmeyer, University of Neuchatel, Switzerland

Background:

Coherent vacuum- and extreme ultraviolet (VUV/XUV) light sources open up new opportunities for science and technology. The most promising technique for table-top sources is femtosecond laser-driven high-harmonic generation (HHG) in gases.

Objective(s):

The goal of this project is ultrahigh precision spectroscopy exploiting a new XUV source. The high harmonics generation is placed directly inside the intra-cavity multi-kilowatt beam of a femtosecond laser that operates at multi-megahertz repetition rates.

Impact:

We will realize high precision XUV spectroscopy experiments that were previously impossible. This should improve our understanding of various areas in technology and science, for example in the area of fundamental tests for quantum electrodynamics (QED) like the ultra-precise measurement of the fine structure constant.

Funding:

36 months (2012-2015)
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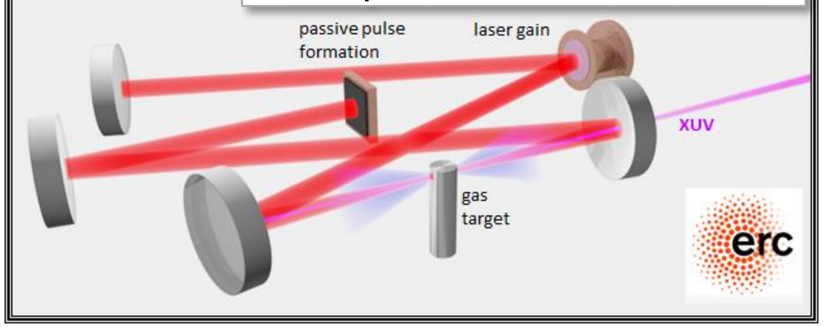
Concept of the intra-laser HHG source

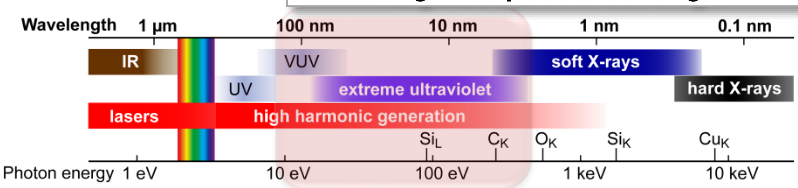


Intra-laser high harmonic generation

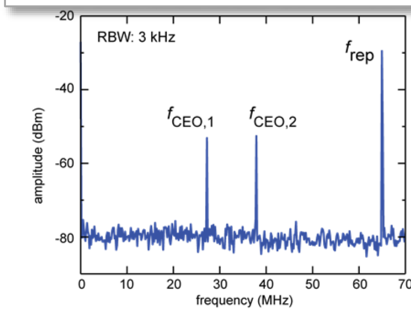
- broad range of harmonics generated simultaneously (UV-XUV)
- spatially and temporally coherent
- Multi-megahertz repetition rates: frequency combs

T. Südmeyer, et al., "Femtosecond laser oscillators for high-field science", *Nature Photonics* **2**, 559 (2008)

Targeted spectral coverage

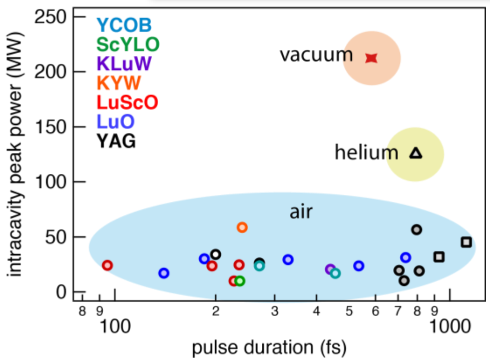


First frequency comb measurement of a thin disk laser demonstrated



C. J. Saraceno, et al., "Self-referenceable frequency comb from an ultrafast thin disk laser," Opt. Express **20** (2012)

Record-high intra-laser peak power levels achieved



C. J. Saraceno, et al., "275 W average output power from a femtosecond thin disk oscillator operated in a vacuum environment," Opt Express **20**, 23535-23541 (2012)

AFRL

Femtosecond laser oscillators for high-field science

Ultrafast laser oscillators have become ubiquitous in science and technology. For many years, however, their pulse energy has been limited to the nanojoule regime. Applications requiring more intense pulses relied on complex amplifier systems, which typically operate at low pulse repetition rates of the order of kilohertz. Recently, the pulse energy of femtosecond laser oscillators has greatly increased, such that some of these experiments can now be driven at multimegahertz repetition rates, which opens promising new avenues for many applications. We review the current state of the art of high-energy femtosecond laser oscillators, in particular mode-locked thin-disk lasers, and discuss their potential to drive high-field science experiments at multimegahertz repetition rates.

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The generation of extremely short laser pulses with the maximum possible pulse energy is an important goal in the research and development of ultrafast lasers. The fast-moving frontier of ever shorter pulse durations[1] has enabled many breakthroughs in science and technology and has been a crucial contributor to two recent Nobel Prizes (femtochemistry by Ahmed Zewail in 1999 and frequency metrology by John Hall and Theodor Hänsch in 2005). Intense femtosecond laser pulses can generate strong electric fields that are comparable to the intra-atomic electric fields. This enables characterization and control of the electronic dynamics that take place inside atoms and molecules by, for example, studying the ionization processes that take place in strong laser fields. High-harmonic generation (HHG) was discovered thanks to such studies[2,3]. High-harmonic generation enables the creation of table-top vacuum-UV (VUV) to soft-X-ray sources, spanning a wavelength range from 100 nm down to 1 nm with femtosecond to attosecond pulses[4,5]. Femtosecond high-energy laser sources are also important for industrial applications. Laser material processing traditionally uses continuous-wave (c.w.) radiation or nanosecond pulses, but is now moving towards picosecond and femtosecond pulses, which provide higher precision and reduced thermal damage[6,7].

Until a few years ago, only complex and expensive laser-amplifier systems were capable of generating ultrashort pulses with an energy sufficiently high for these applications. Femtosecond laser oscillators generated pulses at multimegahertz repetition rates, but their pulse energies were well below the microjoule level. Increasing the pulse energy with amplifier systems introduced several drawbacks in terms of complexity, repetition rate and noise. Although high-repetition-rate seed oscillators were used, typical amplifier systems operated in the kilohertz regime, which was mostly due to the power and repetition-rate limitations of their Q-switched pump lasers.

High-field science experiments, such as the study of ionization processes in strong laser fields, are therefore typically driven at low repetition rates. This reduces the achievable signal-to-noise ratio and increases the time required for many measurements. Amplifier instabilities occurring during the long acquisition time can further degrade the achieved resolution. To reduce the measurement time, the density of the target atoms is usually increased, such that one laser pulse simultaneously ionizes a larger number of atoms. However, in this case space-charge effects (that is, electromagnetic forces between the generated charged particles within the interaction volume) can blur the achievable resolution. Similarly, the low signal-to-noise ratio and long acquisition time caused by the low repetition rate of the driving lasers result in limitations for experiments relying on HHG light sources[8]. The conversion efficiency from the infrared to high-harmonic radiation is typically very low (10^{-6}–10^{-8}). The low average power of the driving amplifier systems therefore results in very low average powers of the generated harmonic radiation, which is well below the milliwatt level. This is a challenge for applications such as extreme-UV (XUV) metrology, high-resolution XUV imaging, and microscopy[8], which all benefit from a higher average photon flux and higher repetition rates. These sources are also crucial for many other applications, for example, surface science and condensed-matter studies using angle-resolved photoemission spectroscopy[9] (ARPES). Space-charge effects have been a limitation for time-dependent measurements in high-temperature superconductors in the VUV regime, and higher repetition rates seem indispensable for precise, time-resolved ARPES.

These reasons provide a strong motivation for developing laser oscillators with high pulse energy at megahertz repetition rates. Since the late 1990s, Ti:sapphire lasers have revolutionized ultrafast science. Recently, it was demonstrated that a semiconductor-saturable-absorber-mirror (SESAM) mode-locked Ti:sapphire laser oscillator with only 0.5 μJ pulse energy can generate sufficiently high peak intensities for multiphoton ionization of noble gases[10].

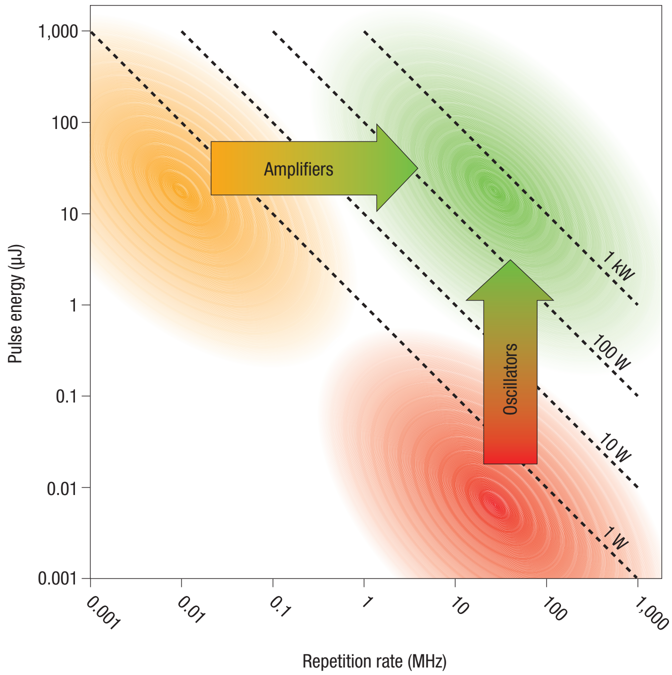

Figure 1 Operation regimes of ultrafast-laser systems according to their typical pulse energy and repetition rate. The diagonal lines correspond to constant average power. Femtosecond lasers can be separated into two different classes, oscillators and amplifiers. Amplifiers typically have substantially increased cost, size and complexity compared with oscillators. Until recently, these two classes of lasers operated in two well separated operation regimes in terms of repetition rate and pulse energy.

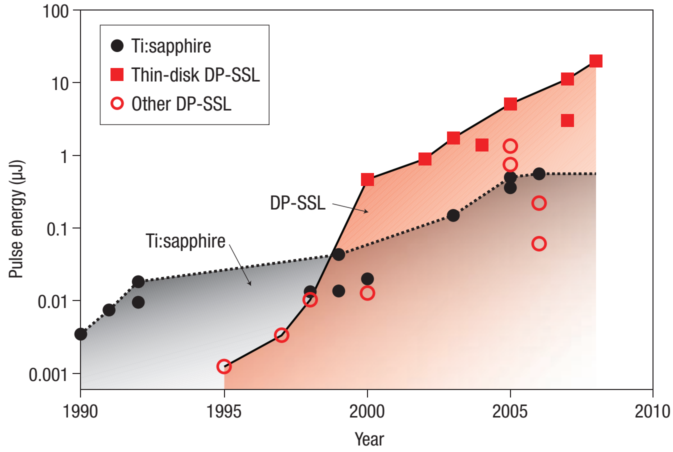

Figure 2 Frontiers in pulse energy from laser oscillators. Maximum pulse energy generated by megahertz femtosecond laser oscillators. Closed black circles: Ti:sapphire lasers; closed red rectangles: thin-disk diode-pumped solid-state lasers (DP-SSL); open red circles: other directly diode-pumped lasers not based on the thin-disk concept. The SESAM-mode-locked thin-disk laser concept defines this frontier and has the potential for further energy scaling by at least one order of magnitude.

Femtosecond diode-pumped thin-disk lasers can generate pulses at megahertz repetition rates with energies beyond the 10-μJ regime[11–14], which is higher than any other mode-locked laser technology. In this article, we discuss the recent progress in diode-pumped solid-state laser oscillators, for which the pulse energy has increased by four orders of magnitude during the past 13 years. At present, the achievable pulse energy is high enough to drive high-field physics experiments at multimegahertz repetition rates. We review this laser technology and discuss its impact on high-field science applications. Using temporally compressed pulses from a thin-disk laser, peak intensities as high as 6×10^{13} W cm^{-2} were recently reported in the first photoelectron-momentum spectroscopy measurements performed with a mode-locked laser oscillator[11,15]. The thin-disk laser technology is power-scalable, and further pulse energy scaling by at least an additional order of magnitude can be anticipated.

HIGH-PULSE-ENERGY HIGH-REPETITION-RATE FEMTOSECOND LASERS

Several research efforts target the development of femtosecond sources that combine high repetition rate and high pulse energy (Fig. 1). Chirped pulse fibre amplifier systems achieved pulse energies as high as 1.8 μJ at 73 MHz and 100 μJ at 900 kHz (refs 16–18). However, these complex systems usually consist of an oscillator, a stretcher and compressor, as well as several amplifier stages. Suppression of higher-order modes in the final-amplifier stage often requires a straight fibre[19], leading to a footprint that is now comparable to standard bulk amplifier systems[20].

The direct generation of energetic femtosecond pulses with a multimegahertz solid-state laser oscillator is a significantly simpler approach to the generation of transform-limited pulses with a high pulse-to-pulse energy stability. Usually, femtosecond oscillators are fundamentally mode-locked, that is, a single pulse propagates inside the optical resonator. The pulse formation is achieved by an intracavity saturable absorber, which introduces lower losses for pulsed compared with c.w. operation of the laser. The most successful passive mode-locking techniques are Kerr lens mode-locking[21] (KLM) and SESAM mode-locking[22,23]. The laser emits a sequence of pulses separated by the roundtrip time, T_R, in the resonator. The pulse energy, E_p, depends on the average power, P_{av}, and the repetition rate, $f_{rep} = 1/T_R$, and follows the relationship $E_p = P_{av}/f_{rep}$. The pulse energy of a mode-locked laser can therefore be increased by increasing the average output power or the length of the oscillator (which directly reduces the pulse repetition rate), or both.

Figure 2 shows the historical evolution of the maximum pulse energy obtained from mode-locked femtosecond solid-state oscillators. In the 1990s, the pulse energy frontier was defined by KLM Ti:sapphire lasers[24,25]. The available pulse energy from femtosecond Ti:sapphire lasers has increased to 0.5 μJ at a pulse repetition rate of up to 6 MHz (refs 10,26, see Fig. 2). At present, thermal issues in the gain material and the need for high-power green pump lasers limit the average power of these oscillators to a few watts. The pulse energy increase was achieved by strongly reducing the laser repetition rate with a Herriott-type multiple-pass cavity[27]. Such a cavity consists of two mirrors, which reflect an incoming laser beam numerous times between each other before it exits the cavity with the same beam parameter. With this approach, a long beam path can be realized inside a compact cavity. Today, the highest pulse energies are generated by diode-pumped solid-state laser oscillators, for which the pulse energies have increased by four orders of magnitude during the past 13 years. In 2000, the first SESAM mode-locked thin-disk laser was demonstrated, which increased the pulse energy of femtosecond diode-pumped solid-state lasers from below 10 nJ to 500 nJ (ref. 28). In contrast to the Ti:sapphire laser, the thin-disk laser is directly diode-pumped and significantly higher average powers are possible with excellent beam quality[29]. At present, mode-locked thin-disk lasers generate up to 80 W of average power[30,31] and pulse energies above 10 μJ (refs 11–14).

In Fig. 3 and Table 1, we compare the operation parameters of different megahertz femtosecond laser oscillators that have high pulse energies.

Two key challenges had to be overcome to increase the energy output of femtosecond oscillators: high-power fundamental transverse-mode operation with broadband gain material at strong thermal load; and stable femtosecond pulse formation at high intracavity pulse energies. The thin-disk laser, which was introduced in 1994 by Giesen *et al.*[29], solves the thermal problems occurring in conventional high-power rod or slab lasers. The gain medium has the shape of a very thin disk, which is mounted directly onto a heat sink and used in reflection. Its typical thickness of 50 μm to 200 μm is small compared with the millimetre-size diameters of the pump and the laser beam. The heat flow is mainly one-dimensional towards the heat sink, which significantly reduces thermal lensing and aberrations. The pump absorption in a single pass through the thin disk is usually weak, but highly efficient operation is achieved by arranging multiple passes through the disk. Stable femtosecond pulse formation is achieved using a SESAM.

All the key components of the passively mode-locked thin-disk laser are used in reflection, and the amount of optical material inside the optical resonator is significantly lower than for other femtosecond oscillators. This is a major advantage for the generation of energetic ultrashort pulses with excellent temporal, spectral and spatial properties, as the pulses are not affected by excessive Kerr nonlinearities inside the resonator. The Kerr nonlinearity is balanced by the appropriate amount of negative group-delay dispersion (GDD), such that transform-limited soliton pulses are formed inside the resonator[32]. This approach is power scalable: the output power can be scaled up by increasing the pump power and the mode areas on both the gain medium and the SESAM by the same factor[28]. This scaling procedure has been verified: the first mode-locked thin-disk laser generated 16 W with a 1.2-mm pump diameter, whereas a thin-disk laser generating 80 W average power uses a pump diameter of 2.8 mm.

While the average power of passively mode-locked thin-disk laser oscillators continuously increased, the pulse energy initially seemed to be limited to values below 2 μJ (refs 30,31). Beyond this level, pulse instabilities and break-up into multiple pulses were observed. In 2006, the Kerr-nonlinearity of the air inside the thin-disk laser cavity was identified as the origin of these instabilities. This additional nonlinearity was too large to be balanced by the negative GDD, which is provided by several dispersive mirrors.

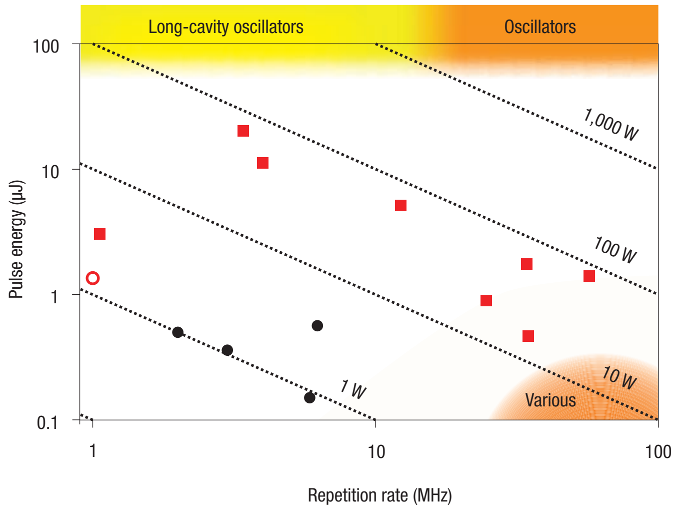

Figure 3 Overview of femtosecond oscillators with high pulse energy. Closed circles: Ti:sapphire lasers; closed rectangles: thin-disk lasers; open circle: other diode-pumped lasers.

Stable soliton mode-locking can either be achieved by reducing the nonlinearity of the atmosphere inside the laser cavity, or by balancing it with additional negative GDD. Covering the Yb:YAG thin-disk laser with an enclosure that was then flooded with helium, which has negligible nonlinearity, eliminated this effect. A pulse energy of 5 μJ at a repetition rate of 12 MHz was achieved, limited by the available power from the pump diodes[33]. Increasing the cavity length using a multiple-pass cavity (Fig. 4a) made it possible to exceed the 10-μJ level at a repetition rate of 4 MHz (refs 11,12). Both pulse envelope and spectrum are well described by an ideal soliton pulse (Fig. 4b,c). This is in contrast to high-power fibre amplifiers, which often suffer from excessive nonlinearities and therefore generate distorted spectra. The output beam was linearly polarized and had a close to ideal Gaussian beam profile (the M^2 value was around 1.1, measured at an average power of 38 W).

Recently, Neuhaus *et al.* demonstrated an Yb:YAG high-power thin-disk laser based on multiple passes through the gain medium[13],

Table 1 Current state of the art of megahertz femtosecond laser oscillators with high pulse energy using either KLM, SESAM mode-locking or cavity dumping.

Laser gain material	ML technique	Wavelength, λ_0	Pulse duration, τ_p	Average power, $P_{av,out}$	Repetition rate, f_{rep}	Pulse energy, E_p	Year	Ref.
Yb:KYW thin disk	SESAM ML, cavity dumped	1,024 nm	680 fs	3.21 W	1.06 MHz	3 μJ	2007	36
Ti:sapphire	KLM, cavity dumped	780 nm	60 fs	0.36 W	0.8 MHz	0.45 μJ	2006	63
Yb:KYW	SESAM ML, cavity dumped	1,040 nm	380 fs	1.35 W	1 MHz	1.35 μJ	2005	64
Yb:glass	SESAM ML, cavity dumped	1,040 nm	250 fs	0.33 W	1.1 MHz	0.3 μJ	2005	65
			470 fs	0.375 W	0.5 MHz	0.75 μJ		
Yb:YAG thin disk	SESAM ML	1,030 nm	810 fs	60 W	34.3 MHz	1.75 μJ	2003	30
Yb:YAG thin disk	SESAM ML	1,030 nm	705 fs	80 W	57 MHz	1.4 μJ	2004	31
Yb:YAG thin disk	SESAM ML	1,030 nm	796 fs	63 W	12.3 MHz	5.1 μJ	2006	33
Yb:KYW thin disk	SESAM ML	1,028 nm	240 fs	22 W	24.6 MHz	0.9 μJ	2002	39
Yb:YAG thin disk	SESAM ML	1,030 nm	730 fs	16.2 W	34.6 MHz	0.47 μJ	2000	28
Yb:YAG thin disk	SESAM ML	1,030 nm	791 fs	45 W	4 MHz	11.25 μJ	2007	11,12
Yb:YAG thin disk	SESAM ML	1,030 nm	811 fs	68 W	3.4 MHz	20.1 μJ	2008	13,14
Yb:KGW	SESAM ML	1,040 nm	134 fs	5.3 W	45 MHz	0.12 μJ	2006	66
		1,040 nm	433 fs	10 W	45 MHz	0.22 μJ		
Ti:sapphire	KLM	800 nm	45 fs	1 W	2 MHz	0.5 μJ	2005	26
		800 nm	40 fs	1.08 W	3 MHz	0.36 μJ		
Ti:sapphire	KLM	800 nm	54 fs	3.5 W	6.23 MHz	0.56 μJ	2006	10
Ti:sapphire	KLM	906 nm	16.5 fs	0.17 W	15 MHz	0.011 μJ	1999	67

ML: mode-locking

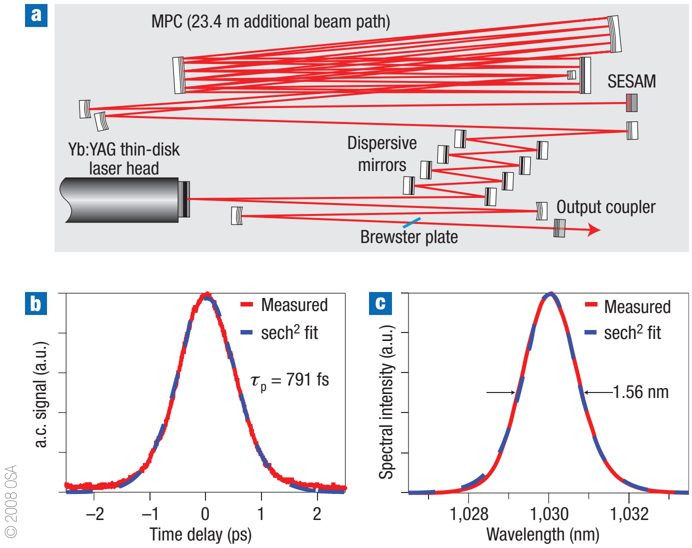

Figure 4 Semiconductor saturable absorber mirror mode-locked, diode-pumped Yb:YAG thin-disk laser generating 11.3 μJ pulse energy at a centre wavelength of 1.03 μm. **a**, Cavity set-up at a pulse repetition rate of 4 MHz using a Herriott-type multiple-pass cavity (MPC), **b**, autocorrelation and, **c**, optical spectrum of the 11.3-μJ output pulses. The average output power was 45 W and the peak power 12.5 MW. τ_p is the pulse duration. Reprinted with permission from ref. 12.

which allowed the output coupling to be increased up to 70%. By using a large amount of negative GDD, operation in air was achieved. Record-high pulse energies up to 20 μJ at a 3.4-MHz repetition rate were obtained with a pulse duration of 811 fs (ref. 14). In c.w. transverse multimode operation, an average power of up to 5.3 kW has been achieved from a single disk with 65% optical-to-optical efficiency[34]. An average output power as high as 360 W with an M^2 of 1.3 has been demonstrated, and even higher power levels will be possible in the future[34,35]. By power-scaling the mode-locked thin-disk lasers, or combining multiple laser heads inside a single resonator, an increase to 500 W average power seems feasible, which would enable a further increase in pulse energy by an order of magnitude.

Cavity dumping of mode-locked oscillators is an alternative approach for the generation of high pulse energies at megahertz repetition rates. In this case, the output coupler of a mode-locked oscillator is replaced by a highly reflective mirror, and a higher pulse energy is built up over a longer time until one pulse is switched out of the cavity with an electro–optical or acousto–optical modulator. A cavity-dumped $Yb:KY(WO_4)_2$ (that is, Yb:KYW) thin-disk laser generated high-energy femtosecond pulses with energies up to 3 μJ at 1 MHz (ref. 36). One of the main challenges is the high nonlinearity in the cavity dumper, which may ultimately limit the achievable pulse energy. Therefore SESAM-mode-locked thin-disk lasers with a multiple-pass cavity are expected to be more suitable for increasing the pulse energy further.

TOWARDS SHORTER PULSE DURATIONS

The optimum pulse duration for driving experiments with femtosecond pulses varies according to the application. Some light–matter interactions, such as material processing, usually depend on the pulse fluence, whereas others depend on the peak intensity. For strong-field applications, peak intensities in the order of 10^{13}–10^{15} W cm^{-2} are required and pulses with tens of megawatts to gigawatts peak power are needed. The pulse duration of mode-locked laser oscillators is limited by the amplification bandwidth of the gain material. The Ti:sapphire gain material has a particularly large optical bandwidth supporting pulse durations of less than 6 fs from low-energy oscillators[37,38]. High-energy Ti:sapphire oscillators operate with longer pulses of around 40 fs as a result of high intracavity nonlinearities and bandwidth limitations[10], achieving peak powers around 10 MW and average powers below 4 W (refs 10,26). Increasing the peak power towards tens of megawatts seems difficult, because of the limitation in average power due to thermal effects and the need for green pump lasers.

So far, mode-locked thin-disk lasers have been realized with only three different gain materials. The highest powers have been obtained using Yb:YAG as the gain medium. Its robustness and high thermal conductivity made it the standard material for high-power applications. The gain bandwidth of Yb:YAG, however, limits the obtainable pulse durations to values near 700 fs in efficient high-power operation. A maximum peak power of 12.5 MW was achieved with the Yb:YAG thin-disk laser discussed above, and the Yb:YAG laser from Neuhaus *et al.*[13] achieved more than 20 MW. Using Yb:KYW with its broader amplification bandwidth, shorter pulses with a duration of 240 fs were obtained at an average power of 22 W (ref. 39). Unfortunately, the strong anisotropy of this material leads to an astigmatism, which makes larger pump spot diameters difficult and therefore limits further increases in average power. The gain material $Yb:Lu_2O_3$ represents a promising alternative to the well established Yb:YAG for high-power laser sources in both c.w. and short-pulse operation[40,41]. This material has a higher thermal conductivity and broader amplification bandwidth, and optical-to-optical efficiencies as high as 72% were achieved in c.w. operation. Recently, the first $Yb:Lu_2O_3$ thin-disk laser was mode-locked, which generated 370-fs pulses with 20.5 W average power and 523-fs pulses with an average power of 24 W. An optical-to-optical efficiency of up to 43% was obtained, which is higher than in previous mode-locked thin-disk lasers[42]. The average power was limited by the available pump power and we expect further power scaling towards several hundreds of watts. The pulses were nearly transform-limited and the output beam was near the diffraction-limit with an M^2 value below 1.1. Other Yb-doped host materials with high potential for achieving shorter pulse duration are $Yb^{3+}:LaSc_3(BO_3)_4$ and $Yb^{3+}:NaGd(WO_4)_2$, that is, Yb:LSB and Yb:NGW, respectively[43,44].

External pulse compression is another approach towards moving into the sub-100-fs regime with existing mode-locked thin-disk oscillators. Pulses with a duration of a few hundred femtoseconds can be efficiently compressed by one additional nonlinear stage, such as a fibre compressor. The nonlinearity of the fibre is used to broaden the optical spectrum, and the resulting chirp is subsequently removed by a dispersive element[45,46]. The multiwatt-average-power regime can be accessed using microstructured large-mode-area fibres and dispersive recompression with a double-pass through a simple prism pair[47]. This method can achieve high compression ratios: pulse shortening of 800-fs pulses from a thin-disk laser to 24 fs increased the peak power from 1.2 MW to 16 MW (ref. 48). The output is linearly polarized and has a nearly diffraction-limited beam quality. This method is very efficient; the compressed pulse typically contains more than 60% of the pulse energy incident on the fibre. The maximum incident pulse energy was limited to approximately 1 μJ as a result of damage to the fibre, which had a 200-μm² effective mode area. The damage threshold is typically proportional to the mode area. Recently, single-mode propagation in fibres with mode areas of 1,000–3,000 μm² was achieved[49,50]. With improved fibres, launching of higher pulse energies, greater than 10 μJ, should become possible, and peak powers of more than 100 MW seem feasible. At the higher pulse energy levels of future thin-disk lasers, we also expect to benefit from hollow-core fibre

compressors and filament compressors, with which damage can be prevented[51].

HIGH-FIELD SCIENCE WITH ULTRAFAST THIN-DISK LASER OSCILLATORS

The suitability of temporally compressed femtosecond pulses from a thin-disk laser for high-field experiments was demonstrated by the first photoelectron imaging spectroscopy[52–54] (PEIS) measurements driven by a multimegahertz laser[11,15]. Photoelectron imaging spectroscopy is a sensitive method for measuring the photoelectron momentum distribution after an ionization process, which can be used for example to identify specific ionization paths and reveal the influence of excited states in the ionization process. It was confirmed that PEIS at multimegahertz excitation results in a high signal-to-noise ratio, short measurement times and a high accuracy. Moreover, these measurements enabled the calibration of the maximum peak intensity at the laser focus[55,56]. At low laser intensities the ionization is dominated by non-resonant multiphoton ionization as well as ionization involving the population of excited electronic states of the atom[53]. As a result of the electric field in the laser focus, the linear ponderomotive shift of high-lying Rydberg states enables resonantly enhanced multiphoton ionization at specific laser intensities[56]. Their appearance in the measured spectra enables a calibration of the peak intensity in the laser focus as a function of the incident pulse energy. A maximum achieved peak intensity of 6×10^{13} W cm^{-2} within an absolute precision better than 10% was determined using 35-fs pulses (Fig. 5).

Many other high-field science applications can equally benefit from a megahertz laser source with high pulse energy. Coincidence measurements of the ion and electron momenta from a multiphoton-ionization process have been used to resolve the long-standing debate about the competitive double-ionization processes based on either re-scattering or sequential double ionization at high and moderate laser intensities[57]. Any coincidence measurement requires that at most one pair of ions and corresponding electrons is generated during each laser pulse.

Another promising area is HHG performed at megahertz repetition rates, which substantially increases the average photon flux for VUV and XUV metrology. So far, megahertz HHG has only been achieved in passive enhancement cavities[58–60]. This method is promising for applications in XUV precision spectroscopy and can, in principle, also be used with high-power mode-locked thin-disk lasers. However, the interaction of light with the gas target occurs inside a high-finesse enhancement cavity, and it is challenging to extract the high-harmonic radiation with high efficiency[61]. An alternative method for enhancing the conversion efficiency is based on quasi-phase-matching techniques in waveguides[62], which would strongly reduce the finesse of an enhancement cavity owing to the high coupling loss into a waveguide. Pulses from high-power femtosecond thin-disk lasers are highly attractive for driving single-pass HHG at multimegahertz pulse repetition rates using quasi-phase-matching waveguides. This concept has potential for the development of a table-top coherent femtosecond VUV or XUV source with high average photon flux. Such multimegahertz VUV and XUV sources would have a high impact in fields as diverse as medicine, biology, chemistry, physics and materials science, enabling applications, such as time-resolved ARPES, VUV and XUV metrology, or high-resolution imaging.

SUMMARY

Femtosecond laser oscillators are becoming more powerful and can access a light–matter interaction regime that previously was limited to complex and expensive amplifier systems. With passively mode-locked thin-disk laser oscillators, pulse energies beyond the 10-μJ level can now be directly generated with tens of watts average power. With such performance, these oscillators can drive high-field physics experiments. These lasers are based on a power-scalable concept and substantially higher average power levels greater than 500 W and pulse energies greater than 100 μJ therefore seem achievable in the near future. Femtosecond thin-disk lasers have a high potential for further applications in high-field science, for example, for table-top coherent multimegahertz VUV and XUV sources using HHG.

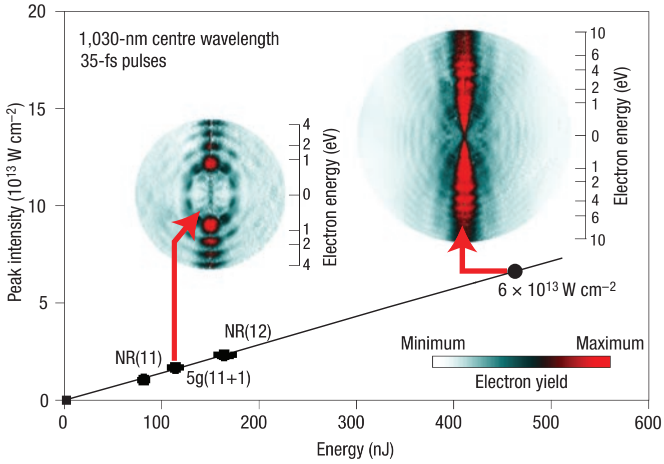

Figure 5 Calibration of the peak intensity by PEIS measurements using a 14-MHz thin-disk laser source[15]. The experimental results were obtained by accumulating the electron signals from more than 10^9 laser pulses. A typical measurement takes 15–70 minutes. To perform this calibration, the appearance of the resonant (11+1)-photon ionization through the 5g state was used (visible in the electron-momentum spectra of xenon at 1.6×10^{13} W cm^{-2}), as well as two calibration points obtained from channel opening of the non-resonant 11- and 12-photon ionization, NR(11) and NR(12), and the zero-energy level[54–56].

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Self-referenceable frequency comb from an ultrafast thin disk laser

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Abstract: We present the first measurement of the carrier envelope offset (CEO) frequency of an ultrafast thin disk laser (TDL). The TDL used for this proof-of-principle experiment was based on the gain material Yb:Lu_2O_3 and delivered 7 W of average power in 142-fs pulses, which is more than two times shorter than previously realized with this material. Using only 65 mW of the output of the laser, we generated a coherent octave-spanning supercontinuum (SC) in a highly nonlinear photonic crystal fiber (PCF). We detected the CEO beat signal using a standard *f*-to-2*f* interferometer, achieving a signal-to-noise ratio of >25 dB (3 kHz resolution bandwidth). The CEO frequency was tunable with the pump current with a slope of 33 kHz/mA. This result opens the door towards high-power frequency combs from unamplified oscillators. Furthermore, it confirms the suitability of these sources for future intralaser extreme nonlinear optics experiments such as high harmonic generation and VUV frequency comb generation from compact sources.

OCIS codes: (140.3580) Lasers, solid-state; (140.4050) Mode-locked lasers; (320.7090) Ultrafast lasers; (320.6629) Supercontinuum generation.

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1. Introduction

Ultrafast thin disk lasers (TDLs), passively modelocked with semiconductor saturable absorber mirrors (SESAMs) [1,2], currently achieve higher pulse energies and average powers than any other modelocked oscillator technology. The thin gain medium of only few 100 µm allows for efficient heat removal and reduced thermal distortions. Using the well-established gain material Yb:YAG in a multi-pass geometry, a pulse energy >40 µJ has been demonstrated with 1.1 ps pulses at an average power of 145 W [3,4]. A comparable average output power level of 141 W with a pulse duration of 738 fs has been demonstrated with one single pass over a disk based on the sesquioxide material Yb:Lu_2O_3 [5].

In principle, the output power of TDLs can be simply scaled up by proportionally increasing the beam diameters on the thin disk gain medium and the SESAM, without excessive increase of nonlinear effects. Therefore, they are excellent candidates for driving experiments requiring high intensities at megahertz repetition rates in systems with the footprint of a low power oscillator [6]. In particular, the high average power levels achievable appear promising to boost the average photon flux in high harmonic generation (HHG) [7,8] and therefore generate a table-top source of vacuum ultraviolet (VUV) and extreme ultraviolet (XUV) radiation. Recently, a record-high average power of 200 µW in the UV was achieved using a passive enhancement cavity seeded by a multi-stage Yb-doped fiber amplifier source operating at MHz repetiton rates [9]. Efficiently driving such highly nonlinear processes requires short pulses durations (<100 fs). Modelocked TDLs were typically limited to pulse durations >200 fs [10] and required pulse compression schemes to reach the sub-100 fs regime [11–13]. Most recently, we have demonstrated for the first time sub-100 fs pulses directly from a modelocked TDL using the mixed sesquioxide material Yb:$LuScO_3$ [14].

Considering these very recent results that confirm that TDLs can directly access the sub-100 fs regime [14] and the carrier envelope offset (CEO) frequency stabilization discussed in this paper, making use of the high intracavity intensity levels inside a TDL to drive extreme nonlinear optics experiments appears very promising. In this approach, which has not yet been demonstrated, one benefits from the high intracavity intensity levels achievable in TDLs to drive for example HHG. Typically, modelocked TDLs operating with one pass over the thin gain medium require low outcoupling coefficients (< 10%). Therefore, they operate at intracavity average powers in the kW regime, pulse energies in the 100 µJ range and peak powers of several tens of MW. For example, the above mentioned femtosecond Yb:Lu_2O_3 TDL with 141 W average output power operated at 1.56 kW intracavity average power and with an intracavity peak power of 31 MW [5], while the high-energy Yb:YAG TDL in [15] already achieved 791 fs pulses with an intracavity energy of 113 µJ at a peak power of 125 MW. Even the recently demonstrated sub-100 fs low-power TDL [14] achieves an intracavity peak power >25 MW and an average power of >200 W, which is already an interesting starting point for first proof-of-principle experiments.

An important range of applications such as VUV/XUV precision spectroscopy on He^+ [16] or even exploring nuclear transitions [17] would benefit from these table-top sources. Very recently, the first demonstration of direct XUV frequency comb spectroscopy was reported [18] at MHz repetition rate using a UV frequency comb driven by a high power fiber based system coupled to a passive enhancement cavity.

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In contrast to passive enhancement cavities [19–22] where the circulating pulse has to match the driving pulses, both pulse formation and laser amplification would be achieved inside the TDL cavity, where the nonlinear process takes place. In addition, there is no need for coherent coupling of the driving pulses, which is a challenging point in passive enhancement cavities. Furthermore, when driving HHG inside a TDL, the circulating pulse can simply adapt to the present nonlinearity. Therefore, bi-stability issues observed in high-finesse passive enhancement cavities due to plasma formation are minimized [23]. Another potential advantage is that different transverse mode profiles can be achieved, for example TEM01, for efficient output coupling of the high harmonics via a hole in a cavity mirror [24]. This is not the case in passive enhancement cavities, where efficient extraction of the UV radiation from these very high finesse cavities is challenging [25,26].

The frequency stability of ultrafast TDLs has not been studied before, which is a key aspect for experiments in the area of high field science and spectroscopy. High power levels are very attractive, because an increase in the average power of frequency combs results in a higher power per mode. So far, stabilized multi-stage fiber chirped pulse amplifier (CPA) systems have reached up to 80 W average power [27]. TDLs can reach similar or higher power levels directly from the oscillator. Prior to the work presented here it was not clear whether pump-induced instabilities could potentially increase the noise level such that a stable frequency comb cannot be realized [28]. TDLs are pumped by high power diodes, which operate in a multimode transverse beam (the fiber-delivered pump beam typically has $M^2 > 100$). Furthermore, they require current drivers operating at several tens of amperes.

Here we investigate for the first time the carrier envelope phase properties of a modelocked TDL. For this task, we realized an Yb:Lu_2O_3 TDL achieving a pulse duration of only 142 fs, which is more than twice shorter than previous TDLs based on this material [29]. These record short pulses enabled the generation of a coherent octave-spanning SC launching less than 1% of the available output power of our TDL directly into a highly nonlinear PCF. The SC was then used in a standard *f*-to-2*f* interferometer [30], enabling us to measure for the first time the CEO frequency beat signal of a modelocked TDL.

2. Yb Lu_2O_3 thin disk laser with short pulse duration

The laser setup used for this experiment is shown in Fig. 1a. The thin disk, used as a folding mirror in the single-mode cavity, consisted of a 150-µm thick, 3%-doped Yb:Lu_2O_3 disk mounted on a 1.4-mm thick diamond heatsink, soldered on a back-cooled copper mount. It had a highly reflective coating for both the pump and laser wavelength on the backside and an antireflective coating for the same spectral range on the front side. Additionally, the disk had a wedge of 0.1° in order to avoid residual reflections which can destabilize modelocked operation. In order to efficiently pump Yb:Lu_2O_3 at its narrow zero-phonon line, we used a volume Bragg grating (VBG) stabilized pump diode [31] emitting at 976 nm in a narrow linewidth $\Delta\lambda < 0.5$ nm. The thin disk module was arranged for 24 passes through the disk enabling an absorption >95% of the pump radiation. Throughout the experiment, we used a pump spot diameter of 1.9 mm.

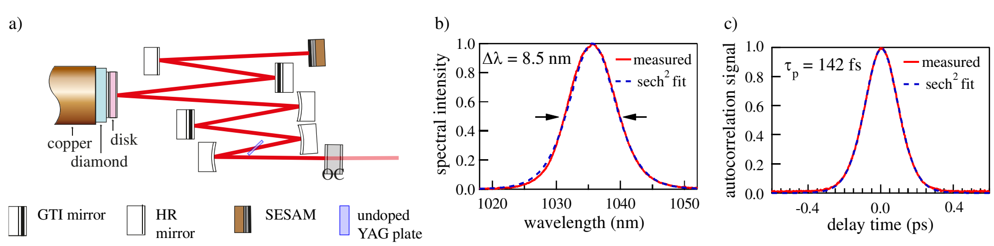

Fig. 1: a) Schematic of the cavity b) Optical spectrum of the pulses at an average power of 7W c) Autocorrelation trace of the corresponding pulses.

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In order to achieve soliton modelocking [32,33], we used two Gires Tournois interferometer (GTI) type mirrors that accounted for 2200 fs^2 of negative dispersion per roundtrip. A 1.5-mm thick uncoated YAG plate, introduced at a focus of ≈200 µm radius accounted for the necessary self-phase modulation (SPM) to balance the negative dispersion in the cavity. Furthermore, it ensures a linearly polarized output. The outcoupling coefficient was 4%. The SESAM used for this experiment was characterized at 1030 nm with 1-ps long pulses. The measurement yielded a saturation fluence F_{sat} = 35 µJ/cm^2, a high modulation depth ΔR = 3.4%, nonsaturable losses ΔR_{ns} = 0.8% and a fast recovery time of $\tau_{1/e}$ = 1.9 ps.

We obtained stable modelocking up to an average power of 7 W. At this average power, pulses as short as 142 fs were obtained with an optical-to-optical efficiency of 15%. This corresponds to a pump power of 47 W. The laser operated at a repetition rate of 64 MHz. The pulses were close to the transform-limit of the spectrum with a time-bandwidth product of 0.34 (Figs. 1 b and c). The corresponding intracavity average power level was 175 W, and the intracavity pulse energy 2.7 µJ. This corresponds to a peak power circulating in the cavity of 17 MW. It is worth noticing that the achieved modelocked optical spectrum (8.5 nm full-width half maximum (FWHM)) is >70% of the available FWHM emission bandwidth of Yb:Lu_2O_3, confirming the large potential of this material also in terms of short pulse generation in the thin disk geometry.

In this experiment, small spot sizes on the disk and the SESAM were used to operate in a relaxed cavity configuration [34,35] and to minimize Q-switching instabilities [36] in the goal of investigating the pulse duration limits of this material. Higher average powers will be reached at these short pulse durations in the near future by using larger disks and laser mode sizes both on the disk and the SESAM.

The SESAM used in this experiment proved crucial for pushing the pulse duration to the limits of the emission bandwidth of this material. In particular, the high modulation depth and fast recovery time have an impact on the stabilization of these short pulses, confirming theoretical predictions of soliton modelocking [32,33]. In this proof-of-principle experiment, we focused on obtaining short pulses and achieved the crucial SESAM parameters in samples with moderate saturation fluence. Future designs will combine these crucial parameters (large modulation depths, fast recovery times) with larger saturation fluences and damage thresholds by using dielectric topcoatings [37], allowing to reach higher power levels.

3. CEO beat detection

We generated a coherent SC in a 1-m long, highly nonlinear PCF using only 65 mW out of the available 7 W of our Yb:Lu_2O_3 TDL. The fiber used is a commercially available highly nonlinear PCF (NKT Photonics A/S, product NL-3.2-945) with a nonlinear parameter γ = 23 $W^{-1}km^{-1}$, and a zero dispersion wavelength of 945 nm. At the laser wavelength of 1034 nm, the fiber exhibits anomalous dispersion of approximately −15.1 ps^2km^{-1}. Considering an estimated coupling efficiency of 50%, the corresponding soliton order launched into the nonlinear fiber is N = 5. According to numerical simulations [38], and recent experiments [39] a soliton order N < 10 is required for the generation of the coherent supercontinuum in this fiber. The short pulses of our TDL enabled the generation of this coherent SC without the need for external pulse compression or amplification. The SC (Fig. 2b) after the PCF covered more than an octave and was launched into a standard f-to-2f interferometer [30] for CEO beat detection. The technical details of the components in the f-to2f interferometer are described in detail in [39]. The CEO beats had a signal-to-noise ratio (SNR) of >25 dB in a resolution bandwidth (RBW) of 3 kHz (Fig. 4c of [39]) and >30 dB in a RBW of 1 kHz. We believe that the achieved SNR ratio is large enough for initial locking tests of the CEO beat, in particular given the high stability of the observed beats. During the time of the experiment (approximately one hour) we did not observe significant frequency excursion or amplitude fluctuations of the CEO beats. Furthermore, significantly better SNR can be achieved by optimizing the laser for low noise performance. It is important to notice that the TDL used for

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this proof-of-principle experiment was built with standard optomechanics, and was not isolated in terms of external vibrations. Furthermore, the pump laser was operated at only 15% of its maximum operation current. Therefore, we believe better performance is achievable by improving the mounting technique, boxing, and pump operation point. Furthermore, optimizing the CEO-beat detection scheme (fiber length, input power level, fiber design, temperature stabilization of the fiber [40], etc...) should also result in an improved SNR.

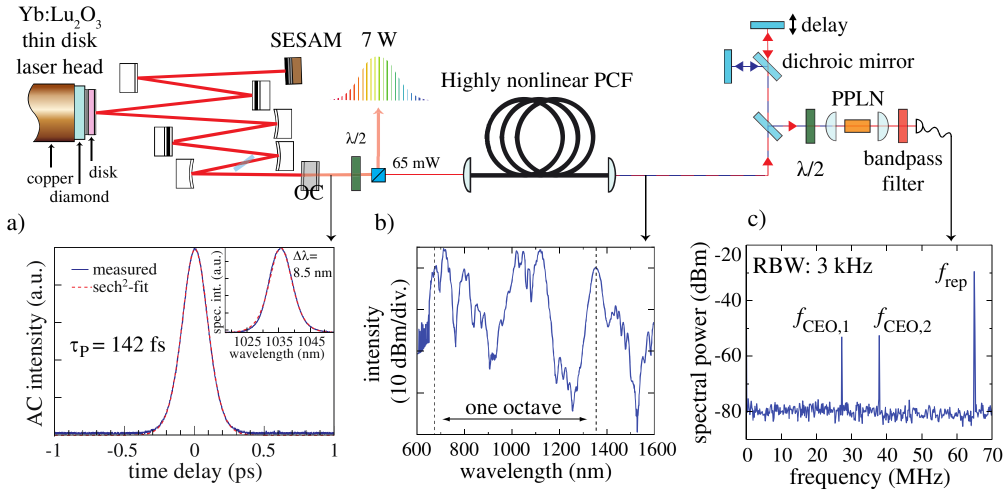

Fig. 2: CEO frequency measurement using a standard f-to-2f interferometer [30]: a) Schematic of the $Yb:Lu_2O_3$ modelocked TDL b) A small fraction of the output power of this laser is enough to generate a coherent SC from a 1-m long highly nonlinear PCF c) The generated SC is launched into a standard f-to-2f interferometer for CEO beat detection.

To investigate the influence of the pulse duration on the detected CEO beats, we increased the pulse duration to 172 fs according to the soliton formula [32] by lowering the pump power and operating at 5.1 W output power. We could still clearly detect the CEO beats at this longer pulse duration, at the expense of a lower SNR (16 dB in 3 kHz RBW). According to numerical simulations [38], recently confirmed experimentally [39] the generation of a coherent SC in a given nonlinear fiber sets a lower limit in terms of pulse duration of the source. For our system, this limit was calculated to be at a pulse duration of approximately 180 fs, which seems to be in accordance with our experiment. Further investigations will target to confirm this limit in pulse duration experimentally. In our experiment, further lengthening of the pulse duration was not possible without breaking into the Q-switched modelocking regime.

The CEO beat frequency was tunable by the pump current, with a slope of approximately 33 kHz/mA. This mechanism can be used for electronic stabilization of the CEO frequency to an external reference. It is worth emphasizing that CEO detection was possible in spite of the strongly multimode pumping scheme of TDLs, usually associated with a high noise level. This seems to indicate that systems such as the one presented in [5] with an external passive pulse compression stage to reach the necessary short pulse duration for CEO detection would already be suitable to achieve >100-W-level stabilized frequency combs.

4. Conclusion and outlook

Our experimental results represent the first confirmation of the potential of modelocked TDLs as high-power stabilized frequency combs. Pulses as short as 142 fs were obtained with the sesquioxide material $Yb:Lu_2O_3$, which has already demonstrated its suitability for high power operation in the thin disk geometry [5]. This proves the potential of this material also in terms

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of high power short pulse generation. Higher output powers will be achieved in the near future at these short pulse durations by using larger disks, increased mode areas and by designing SESAMs with high modulation depths and fast recovery times such as the one used in this experiment, but higher saturation fluences, lower two-photon absorption effects and higher damage thresholds [37]. We expect to reach more than 100 W output power and kW intracavity levels from such a source with sub-100 fs pulse duration in the near future.

The demonstrated Yb:Lu_2O_3 TDL with 142-fs pulses enabled the first CEO frequency beat measurement of a modelocked TDL, which was performed without any external amplification or pulse compression using a standard f-to-$2f$ interferometer [30]. This measurement established a sufficiently large SNR for future stabilization of the laser system with the pump current. Therefore, high-power stabilized frequency combs in the 100 W range from unamplified laser oscillators appear feasible in the near future. These results further increase our confidence that TDLs are ideal candidates for megahertz intracavity nonlinear optics experiments, such as high harmonic generation for future compact XUV/VUV sources.

Acknowledgments

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Towards efficient high harmonic generation inside an ultrafast thin disk laser

Thomas Südmeyer, Physics Department, University of Neuchatel

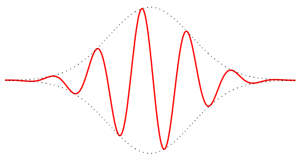
Research activities

Compact rubidium cell frequency standards
Miniature atomic frequency standards based on micro-fabrication
Continuous cesium fountain clock
QCLs, mid-IR detectors
Novel frequency combs and applications, ERC MEGA-XUV

Ultrafast lasers

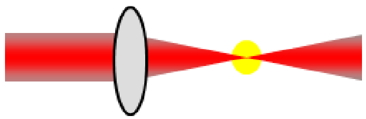
… generate coherent light pulses with pico- or femtosecond duration

Access ultrashort time scales



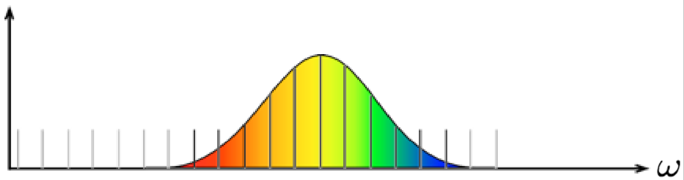
⇨ **Observe and use fast dynamics**
- understand chemical reaction dynamics
- fast communication
- …

Concentrate in time and space



⇨ **Achieve extremely high intensities**
- material processing
- multi-photon biomedical imaging
- high harmonic generation, …

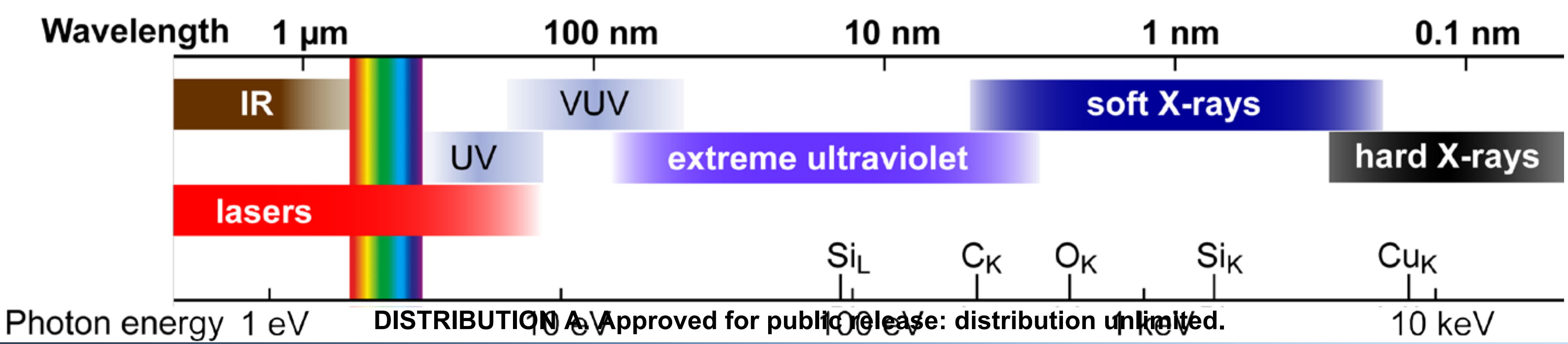
Broad optical spectrum



⇨ **Generate ultrastable frequency combs**
- high precision spectroscopy
- optical clocks
- …

… but limited to IR-visible-UV : no standard lasers at <150 nm

- Observe smaller features
- Write smaller patterns
- Understand dynamics in nanostructures
- Elemental sensitivity (core level e^{-})
- Frequency combs in the VUV/XUV

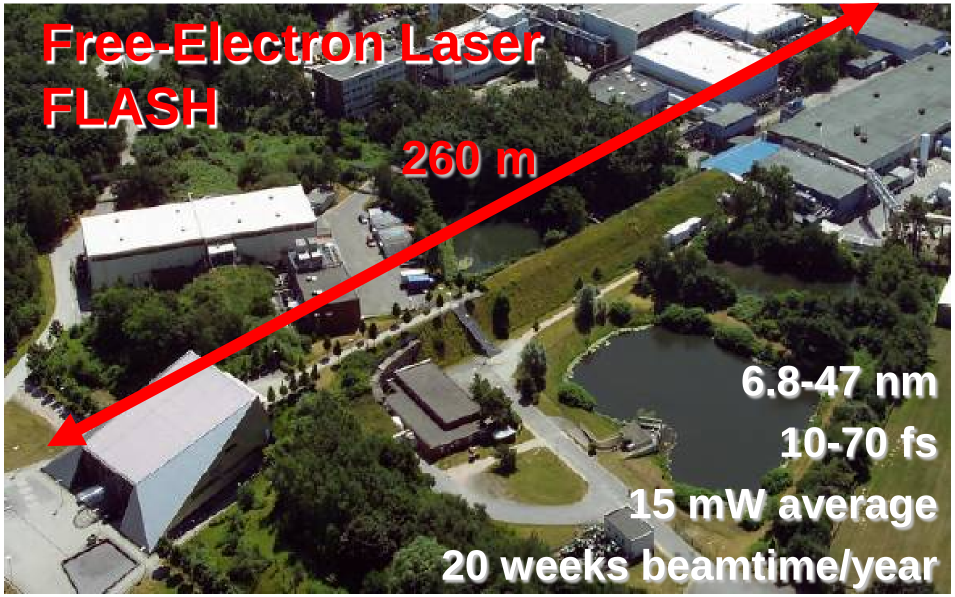


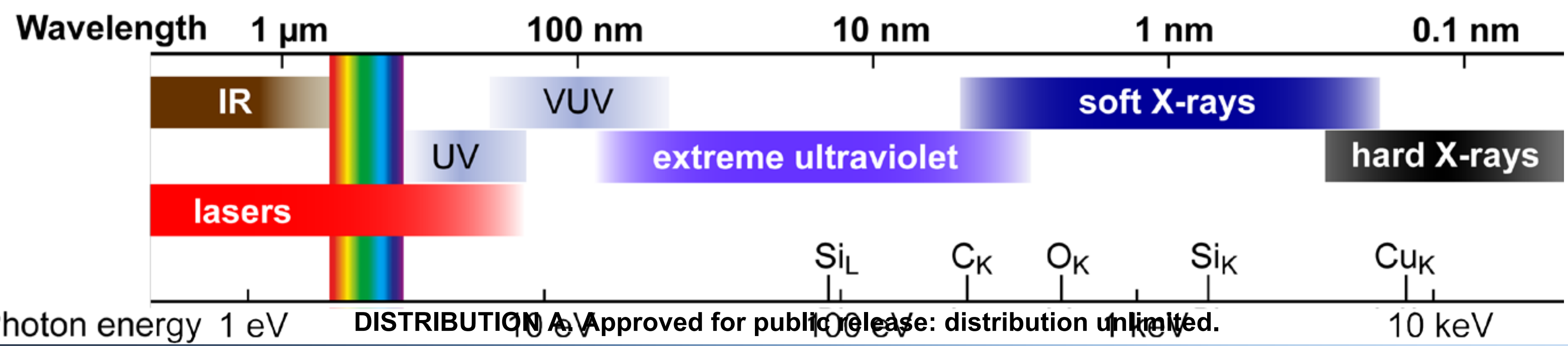
How to generate coherent XUV light?

- Observe smaller features
- Write smaller patterns
- Understand dynamics in nanostructures
- Elemental sensitivity (core level e$^-$)
- Frequency combs in the VUV/XUV

Accelerator-based light sources

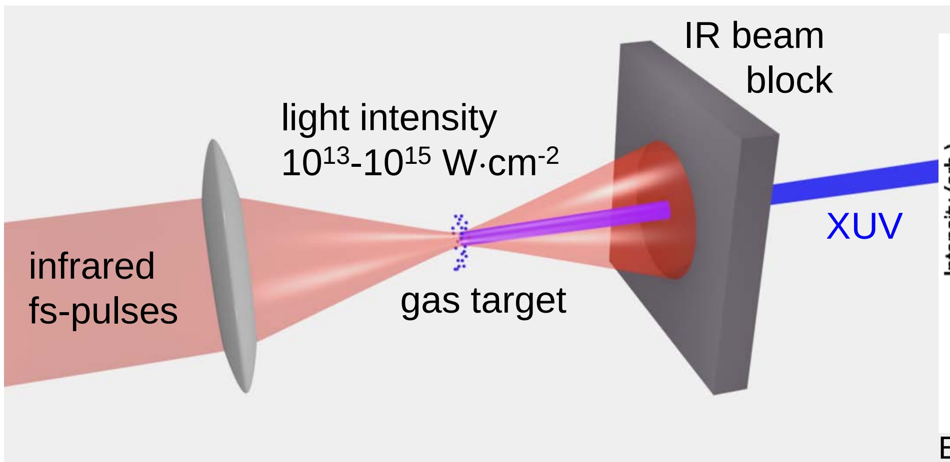
- extremely bright, widely tunable
- but: large, expensive, limited access

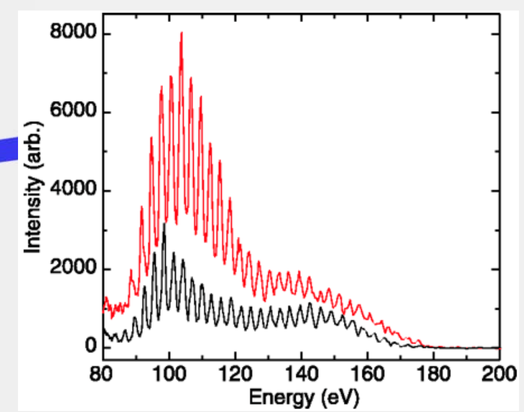




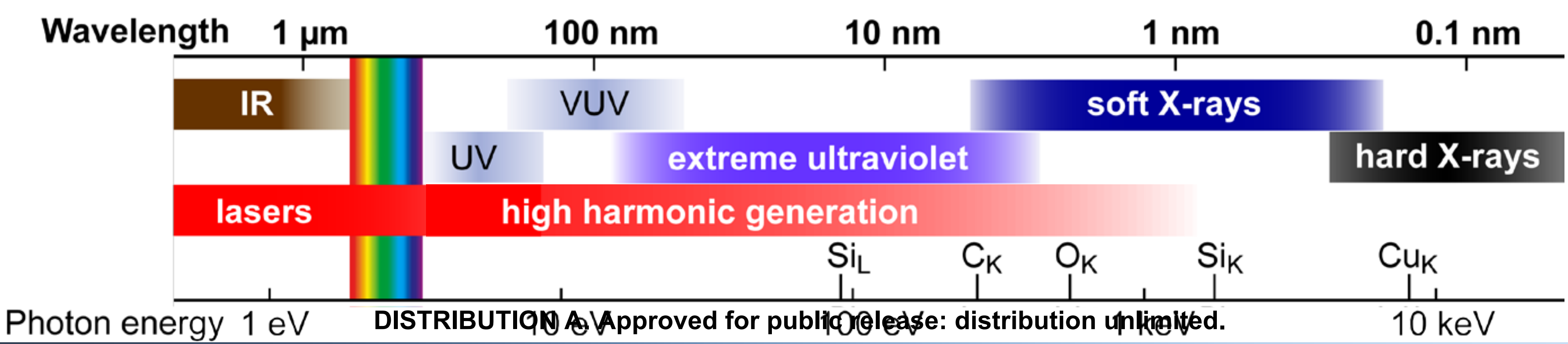
Extreme nonlinear up-conversion of IR fs-pulses in gas target

- broad range of harmonics generated simultaneously (UV-XUV)
- spatially and temporally coherent
- attosecond duration





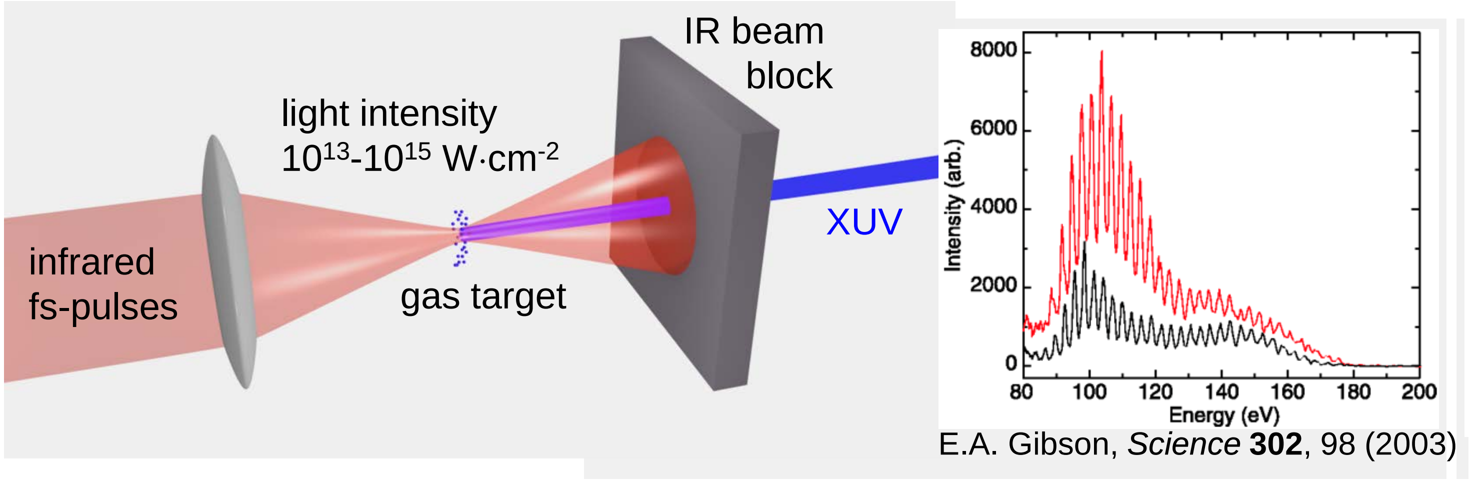
E.A. Gibson, *Science* **302**, 98 (2003)



HHG: bringing coherent XUV light to the lab

Extreme nonlinear up-conversion of IR fs-pulses in gas target

- broad range of harmonics generated simultaneously (UV-XUV)
- spatially and temporally coherent
- attosecond duration



E.A. Gibson, *Science* **302**, 98 (2003)

Limitations

- conversion efficiency 10^{-8} to 10^{-6} ⇨ **flux too low for many applications**
- typical fs-amplifier: 10 W, 1 kHz ⇨ **kHz repetition rate limits usefulness**

Increasing HHG flux and repetition rate

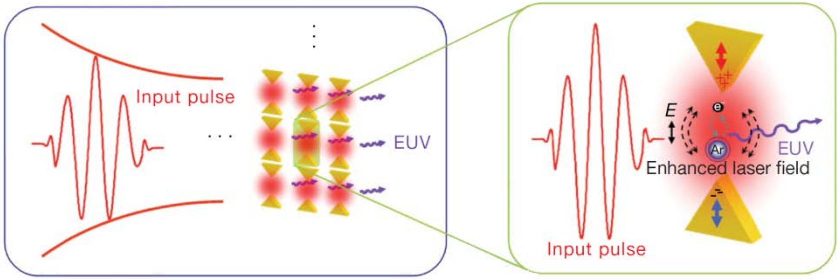
Mainstream approach

- engineer bigger & more powerful femtosecond amplifier systems

… but: future kW-systems suitable for HHG experiments?
reliability? noise?

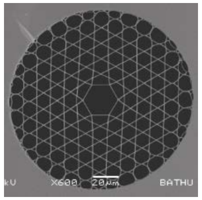
Resonant plasmon field enhancement[#1]

- high repetition rate: 75 MHz
- Diffraction pattern imprinted on high harmonic radiation



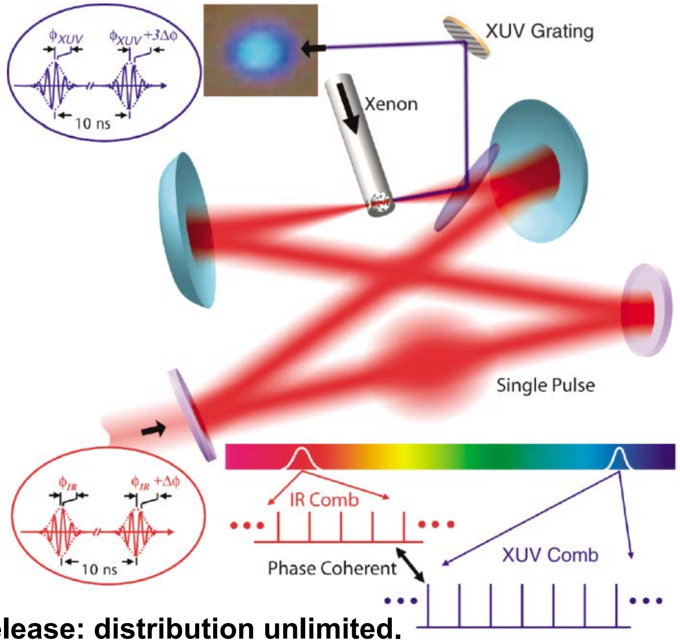
HHG in hollow-core photonic crystal fiber [#2]

- >300 MW peak power in HC-PCF possible
- 200 nJ HHG treshold for 30-fs pulses
- so far only low efficiency ($2\cdot10^{-9}$)



HHG in enhancement cavities[#3,4]

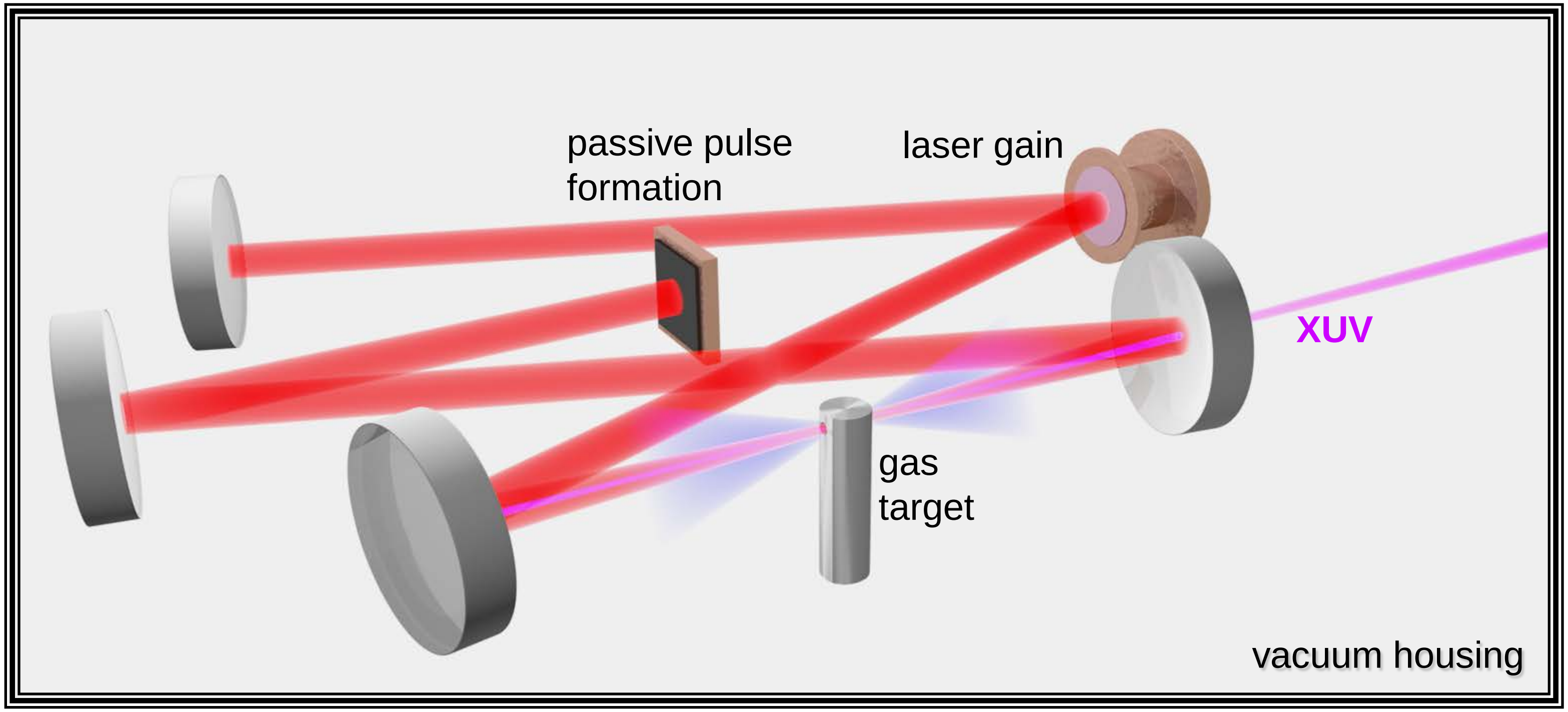
- high enhancement factors >1000 feasible
- > 10 kW intracavity average power achieved
- beam extraction is difficult
- coherent coupling of fs-pulses from laser system required



[#1] Kim, et al., Nature 453, 757 (2008)
[#2] Heckl, et al., Appl. Phys. B 97, 369-373 (2009)
[#3] Jones, et al., Phys Rev. Lett. 94, 193201 (2005)
[#4] Gohle, et al., Nature 436, 234 (2005)

Key idea:

- "recycle" unconverted light inside MHz femtosecond laser resonator
- HHG interaction at kW, but laser gain only has to compensate moderate losses
- in contrast to external enhancement cavities (1,2):
  no limitation by coherent coupling and need for low-losses



1) R. Jason Jones, et al.,"Phase-Coherent Frequency Combs in the Vacuum Ultraviolet via High-Harmonic Generation inside a Femtosecond Enhancement Cavity," Phys. Rev. Lett. **94**, 193201 (2005).
2) C. Gohle, et al., "A frequency comb in the extreme ultraviolet," Nature **436**, 234-237 (2005).

Challenges

High average power

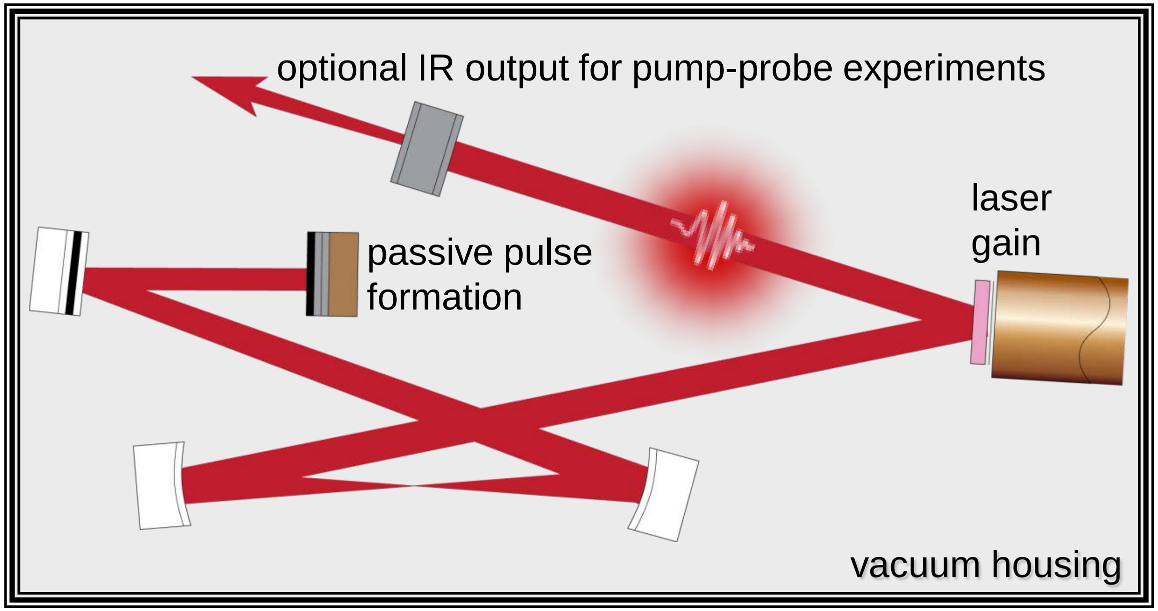
- >10 kW average power, >1 GW peak power feasible?
- Thermal aberrations? Damage?

Stable fs soliton pulse formation [#1]

- Damage of saturable absorber?
- CEP stabilization possible?

Kerr-nonlinearities

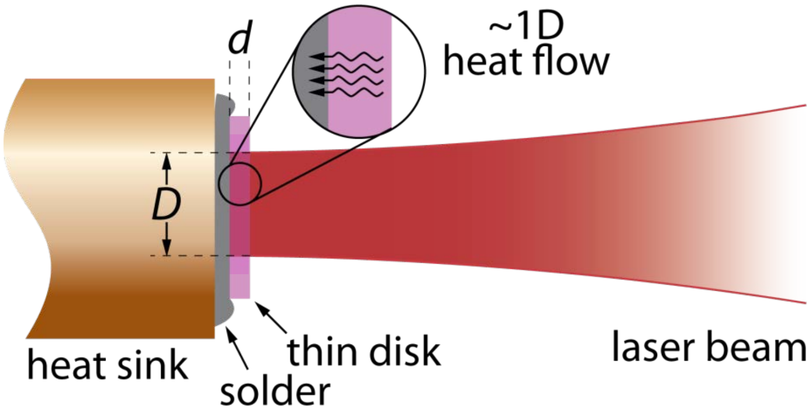
- Self-focussing and damage?

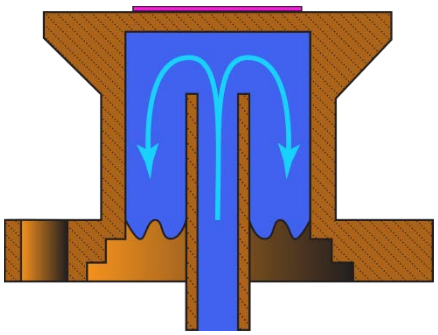


[#1] F. X. Kärtner, and U. Keller, *Opt. Lett.* 20, 16 (1995) and R. Paschotta and U. Keller, *Appl. Phys. B* 73, 653 (2001)

Challenge: Fundamental mode operation despite high thermal load

- Suitable gain material (good optical quality, high thermal conductivity)
- Suitable gain geometry (efficient heat removal)





Thin disk laser

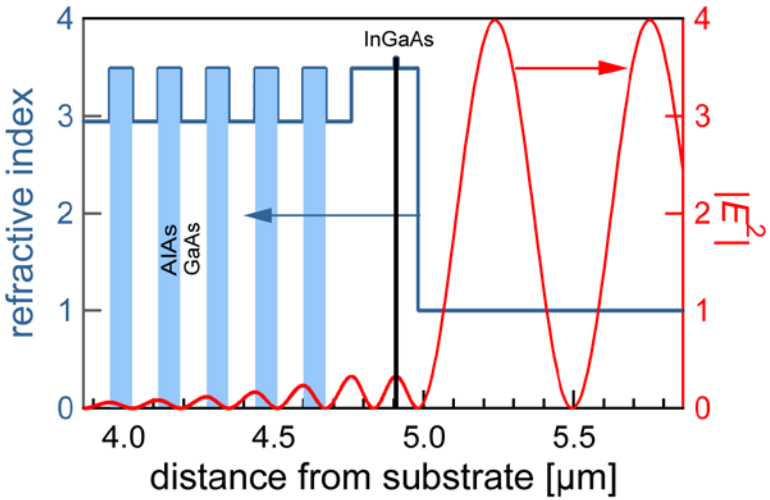
A. Giesen, et al., *Appl. Phys. B* **58**, 365 (1994)

- Efficient heat removal through back side
- Power scalable by increase of mode diameter *D* (constant intensities)
- 1D longitudinal heat flow → reduced thermal lensing

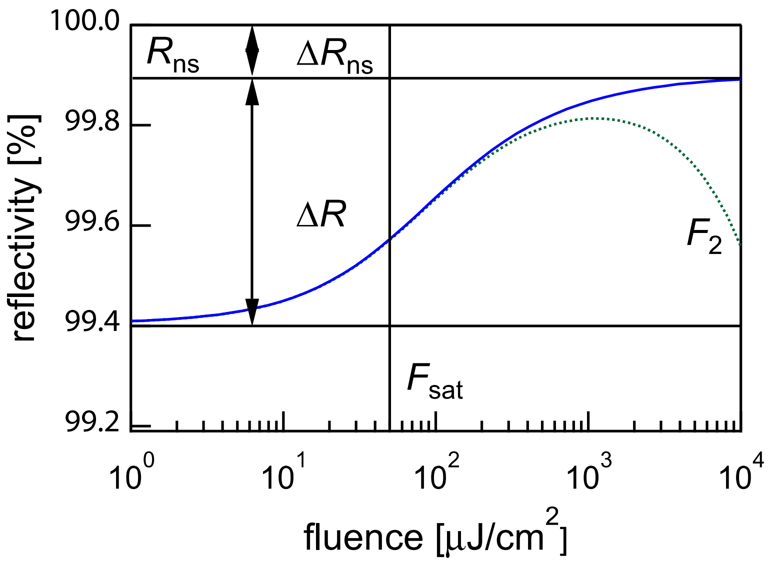
DISTRIBUTION A. Approved for public release: distribution unlimited.

SEmiconductor Saturable Absorber Mirror (SESAM) for passive mode-locking
U. Keller, et al., *IEEE J. Sel. Top. Quant.* 2, 435 (1996)

SESAM structure



Nonlinear reflectivity of a SESAM

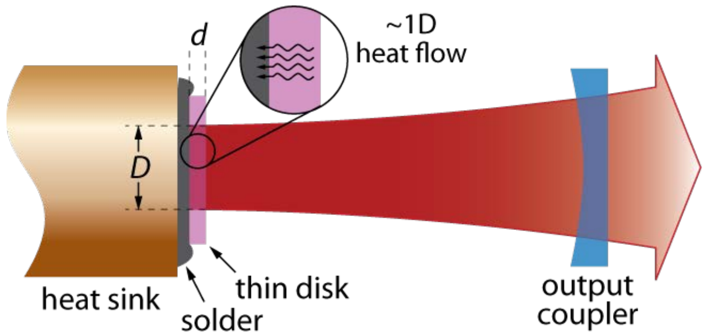


Advantages

- Widely tunable absorber parameter
- Reliable and self-starting passive mode-locking
- High damage threshold (> 100 mJ/cm^2 for optimized design)

The passively mode-locked thin disk laser

Thin disk laser

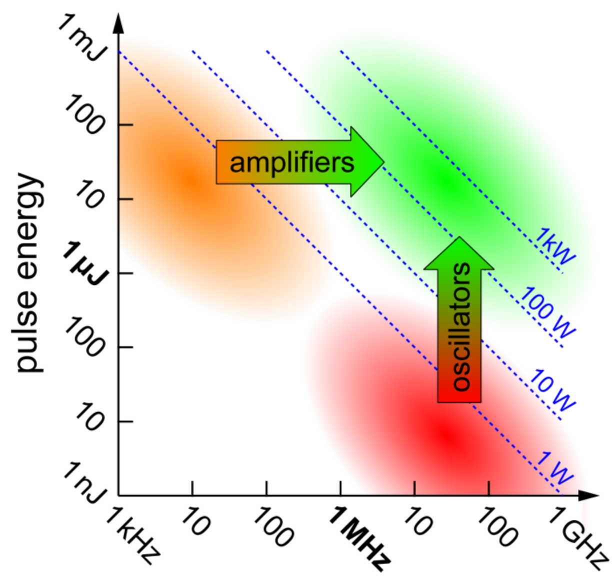


+

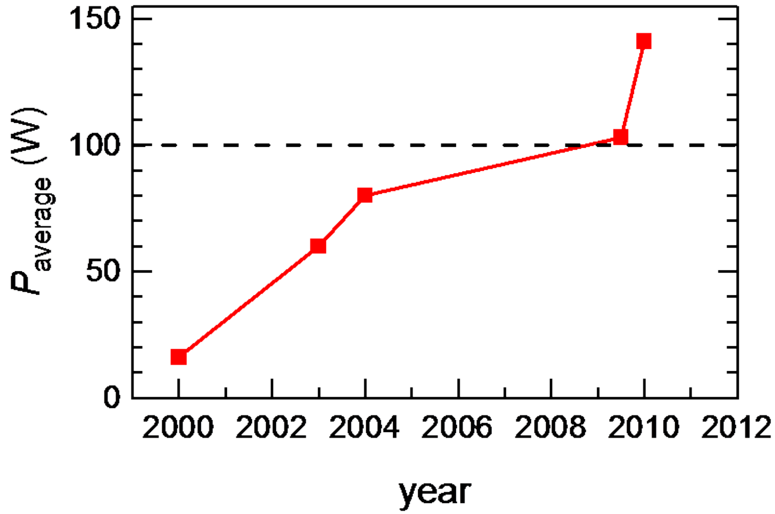
SESAM

semiconductor saturable absorber mirror

A power scalable concept: Scale output power
by equally increasing the pump power and mode sizes on disk and SESAM.



Maximum average power of fs-oscillators



Challenges
- high average power
- stable fs pulse formation
- Kerr-nonlinearities

C. R. E. Baer, C. Kränkel, C. J. Saraceno, O. H. Heckl, M. Golling, R- Peters, K. Petermann, T. Südmeyer, G. Huber, and U. Keller,“Femtosecond laser oscillators for high-field science”, Opt. Exp. 19, (2011)

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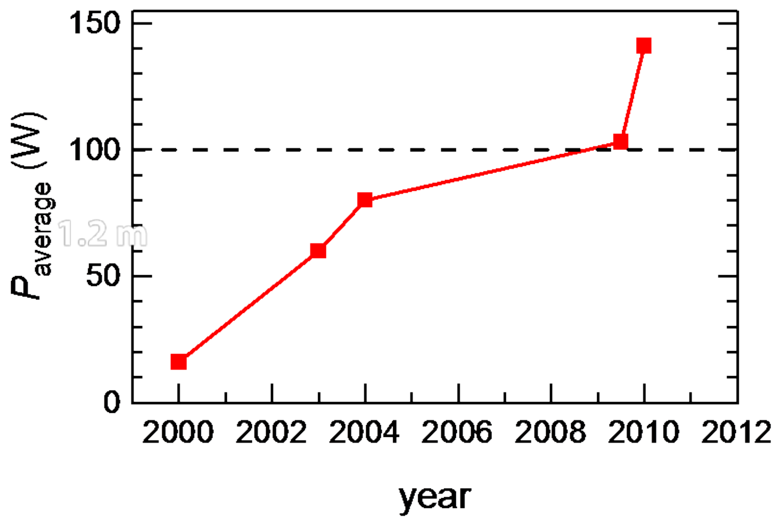
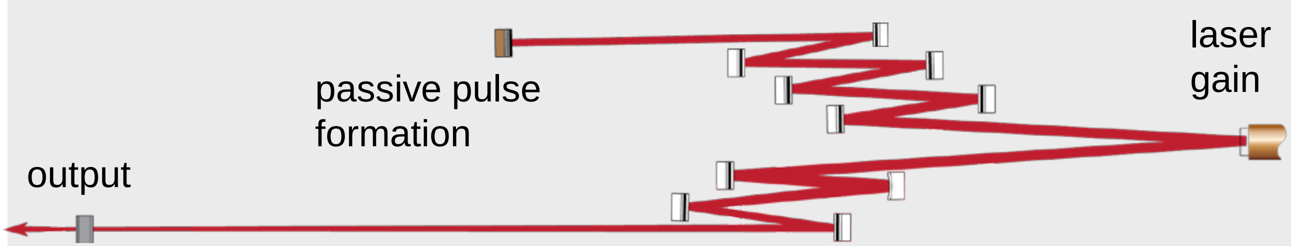


Challenges
- high average power
- stable fs pulse formation
- Kerr-nonlinearities

High power thin disk laser:

140 W, 740 fs, 60 MHz



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High power femtosecond laser oscillators

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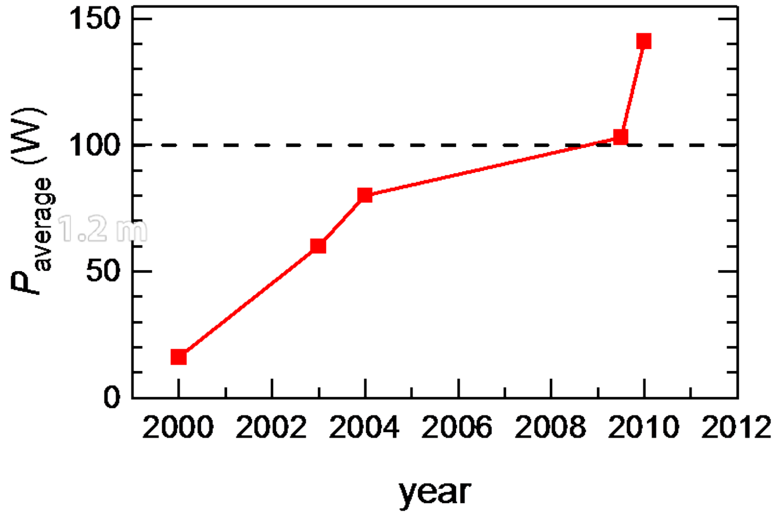
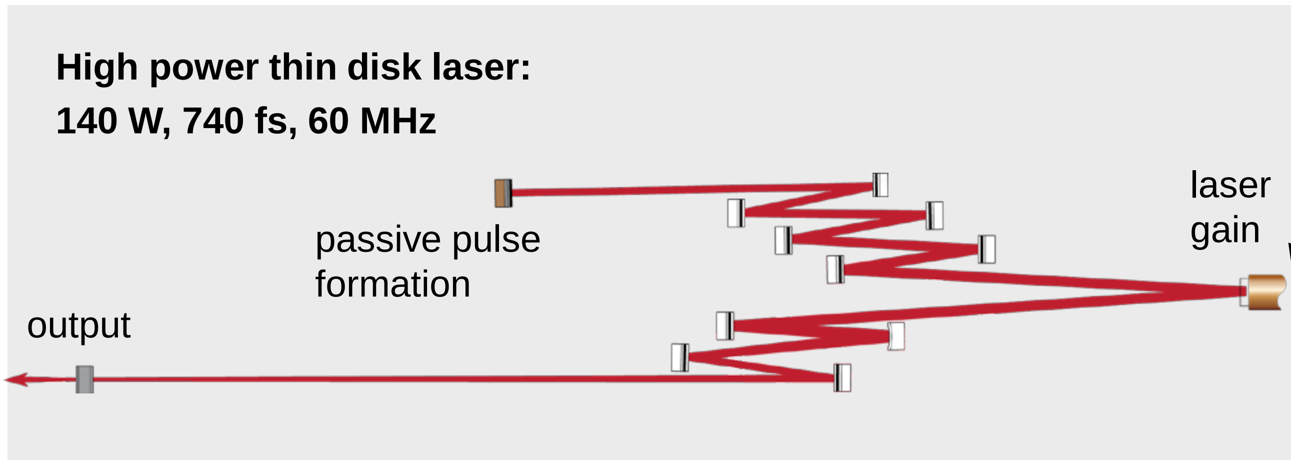
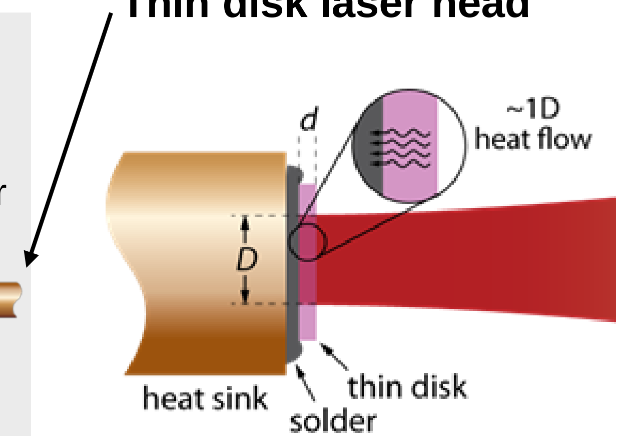


Challenges

- ✓ high average power
- stable fs pulse formation
- Kerr-nonlinearities



Thin disk laser head



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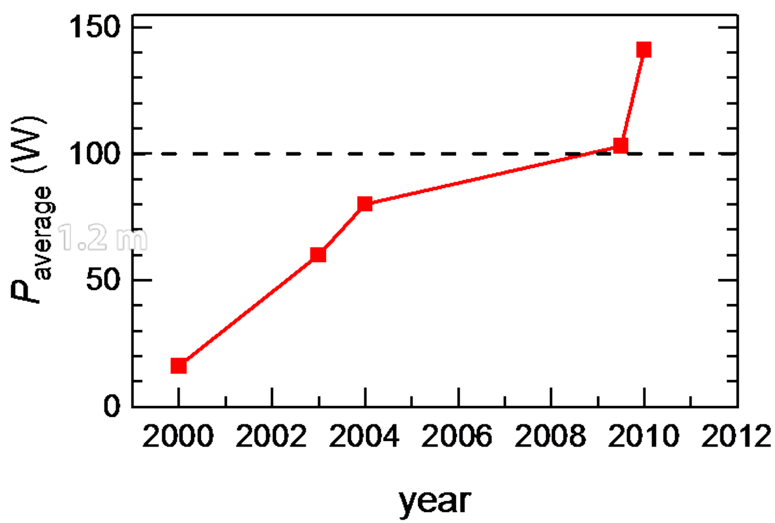
High power femtosecond laser oscillators

Maximum average power of fs-oscillators



Challenges

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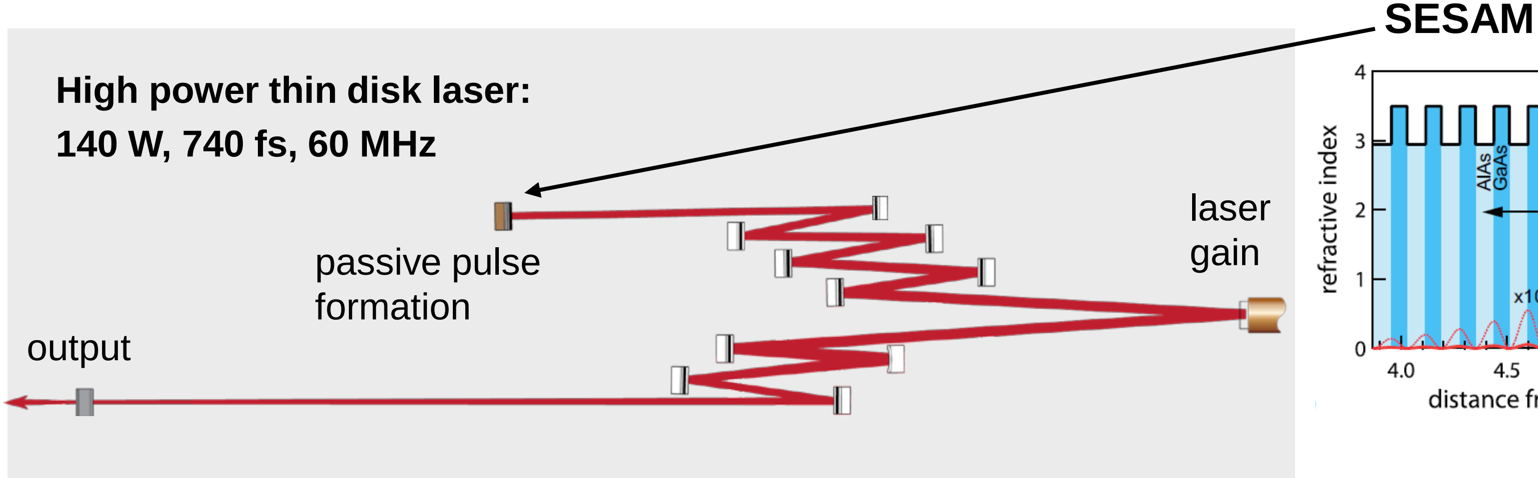

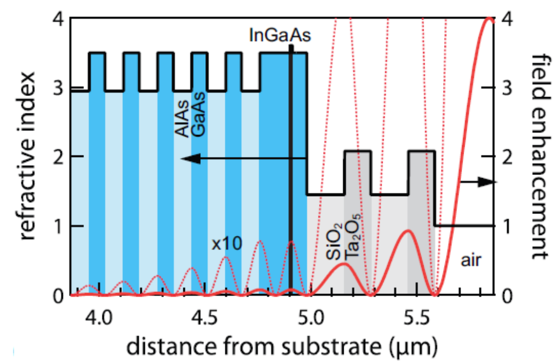

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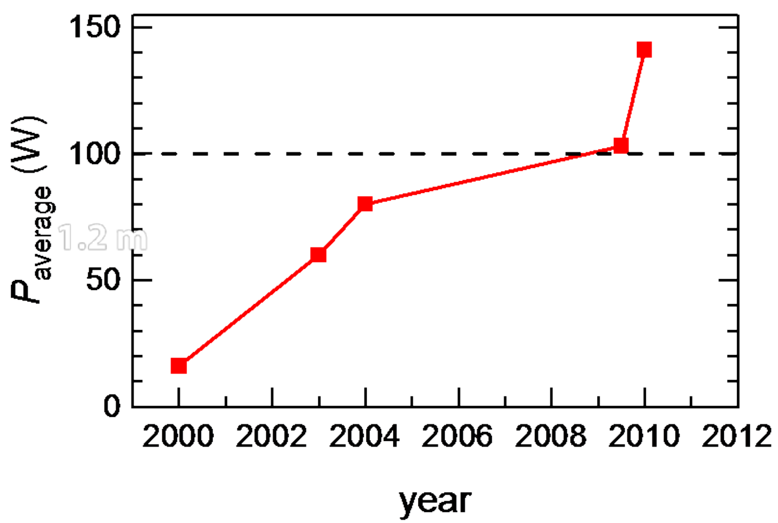
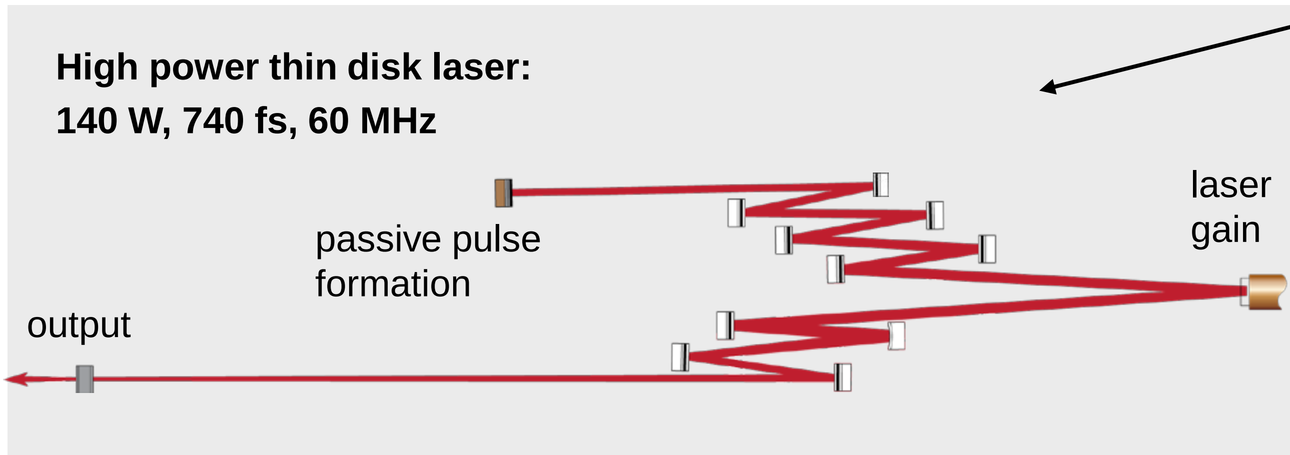


Challenges

- high average power
- stable fs pulse formation
- Kerr-nonlinearities

All components reflective:

only suitable high power fs-oscillator concept



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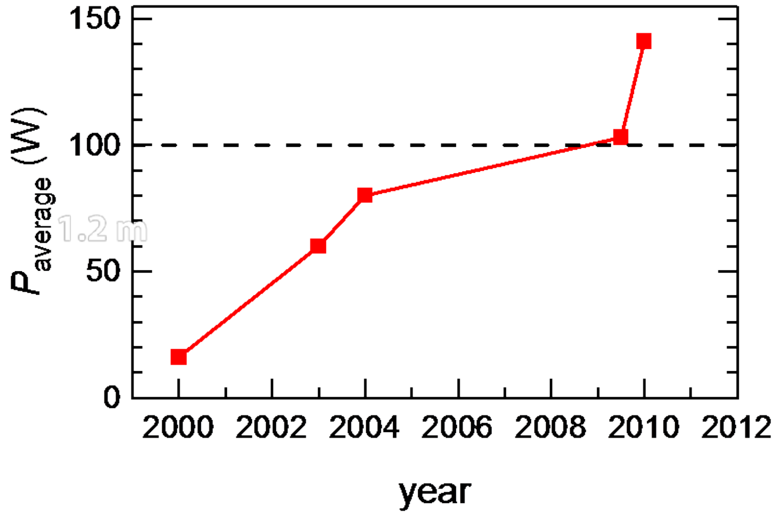
High power femtosecond laser oscillators

Maximum average power of fs-oscillators



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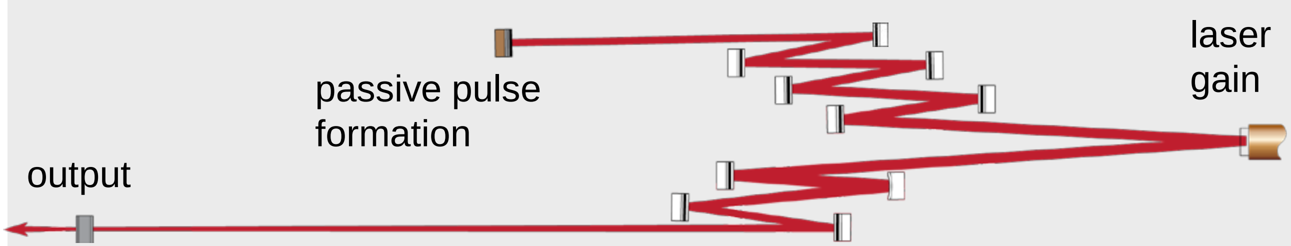

compact setup:
1.2 m x 0.3 m

intracavity power:
1.4 kW

C. R. E. Baer, C. Kränkel, C. J. Saraceno, O. H. Heckl, M. Golling, R- Peters, K. Petermann, T. Südmeyer, G. Huber, and U. Keller,“Femtosecond laser oscillators for high-field science”, Opt. Exp. 19, (2011)

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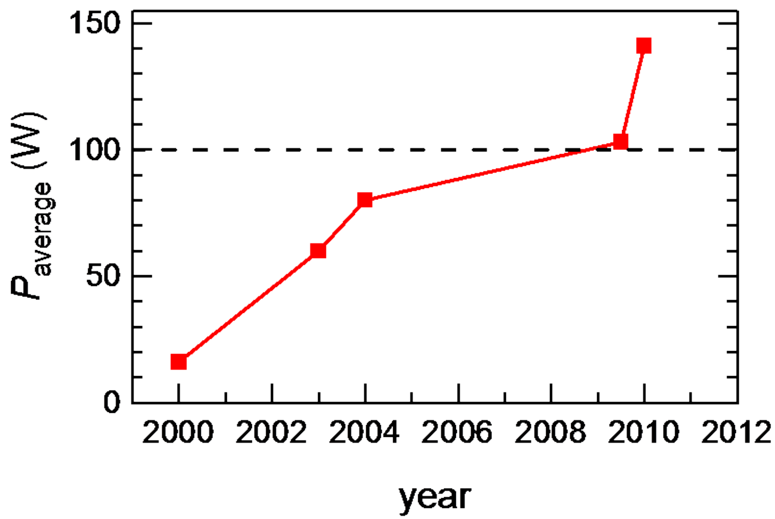

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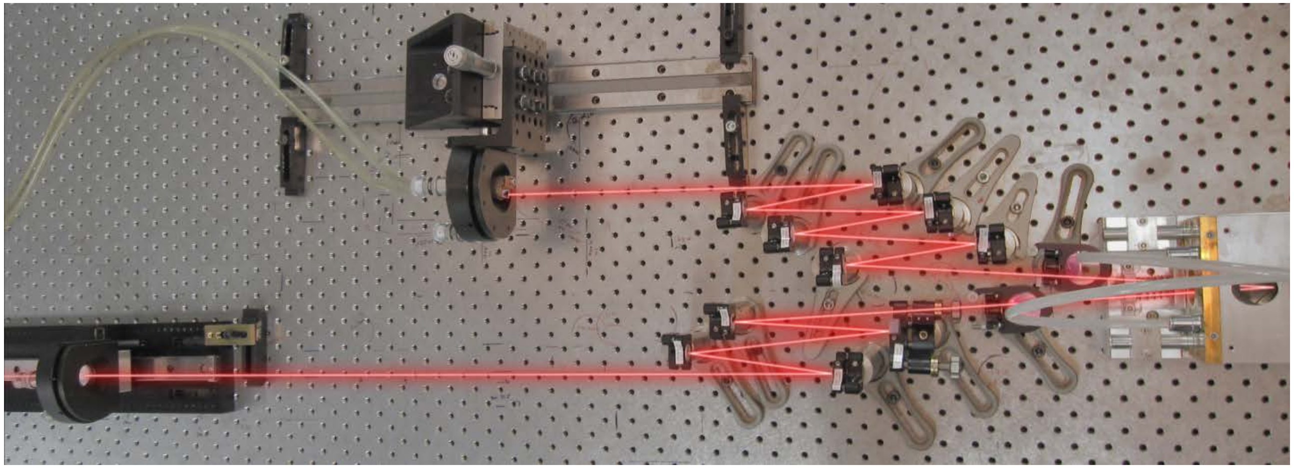

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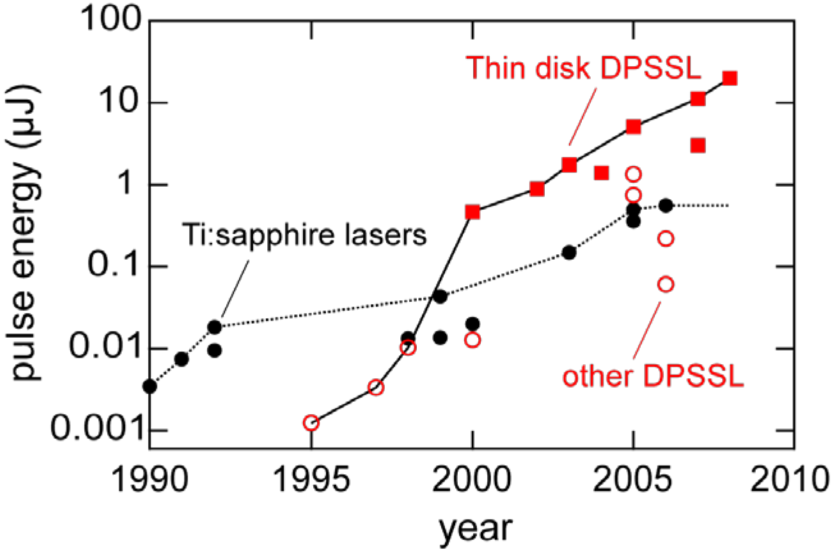
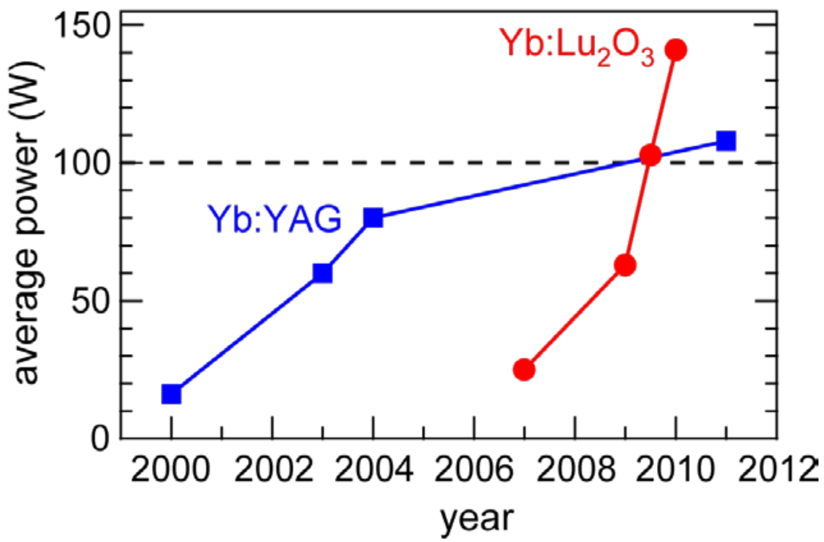
C. R. E. Baer, C. Kränkel, C. J. Saraceno, O. H. Heckl, M. Golling, R- Peters, K. Petermann, T. Südmeyer, G. Huber, and U. Keller,"Femtosecond laser oscillators for high-field science" Opt. Exp. 19, (2011)

High average power and pulse energy

Ultrafast thin disk lasers:
highest pulse energies and average powers
of any modelocked oscillator technology





pulse energy >30 µJ

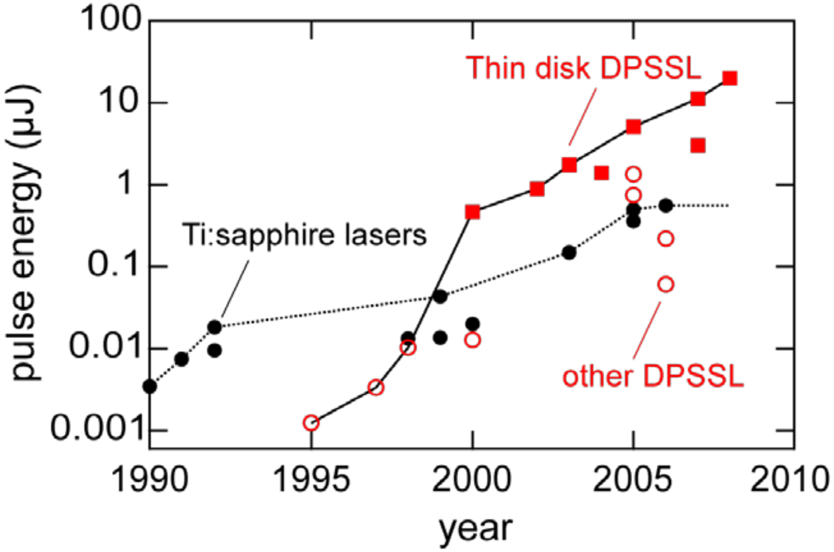
Bauer, Schättiger, Kleinbauer, Sutter, Killi, Dekorsy, ASSP, Istanbul, ATuC2 (2011)

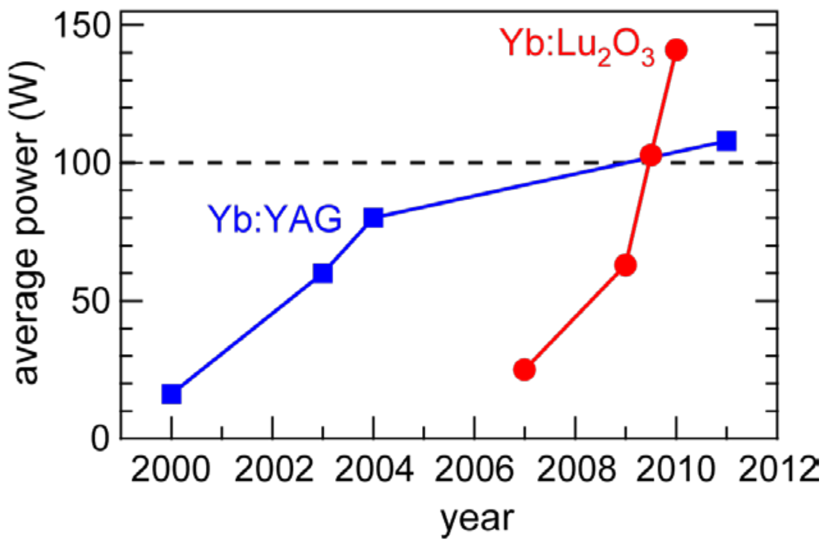
average power >140 W

Baer, Kränkel, Saraceno, Heckl, Golling, Peters, Petermann, Südmeyer, Huber, Keller, Optics Letters 35, 2302 (2010)

T. Südmeyer, et al., "Femtosecond laser oscillators for high-field science", *Nature Photonics* **2**, 559 (2008)

Ultrafast thin disk lasers:
highest pulse energies and average powers
of any modelocked oscillator technology





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Bauer, Schättiger, Kleinbauer, Sutter, Killi, Dekorsy, ASSP, Istanbul, ATuC2 (2011)

duration 1040 fs

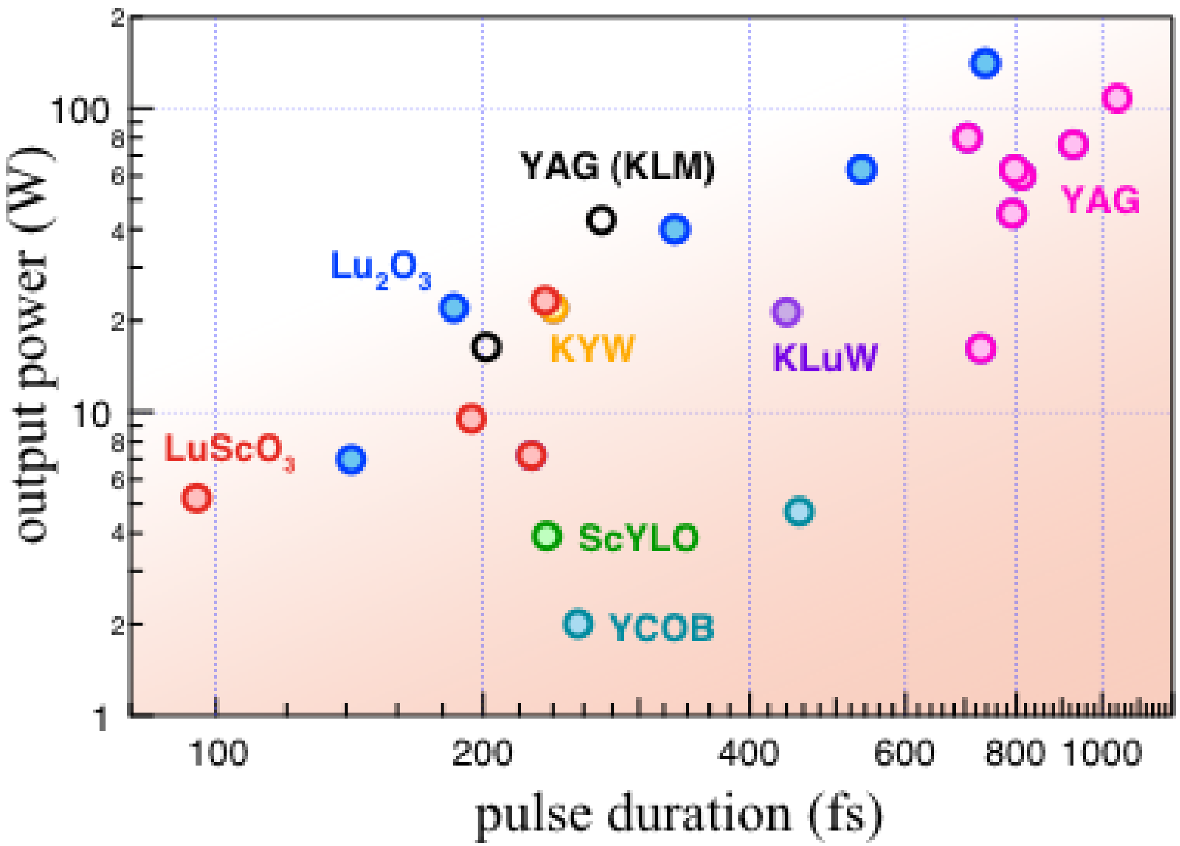
average power >140 W

Baer, Kränkel, Saraceno, Heckl, Golling, Peters, Petermann, Südmeyer, Huber, Keller, Optics Letters 35, 2302 (2010)

duration 738 fs

T. Südmeyer, et al., "Femtosecond laser oscillators for high-field science", *Nature Photonics* **2**, 559 (2008)

Short pulses



Very recent results:

Yb:LuO TDL: achieved 142 fs duration

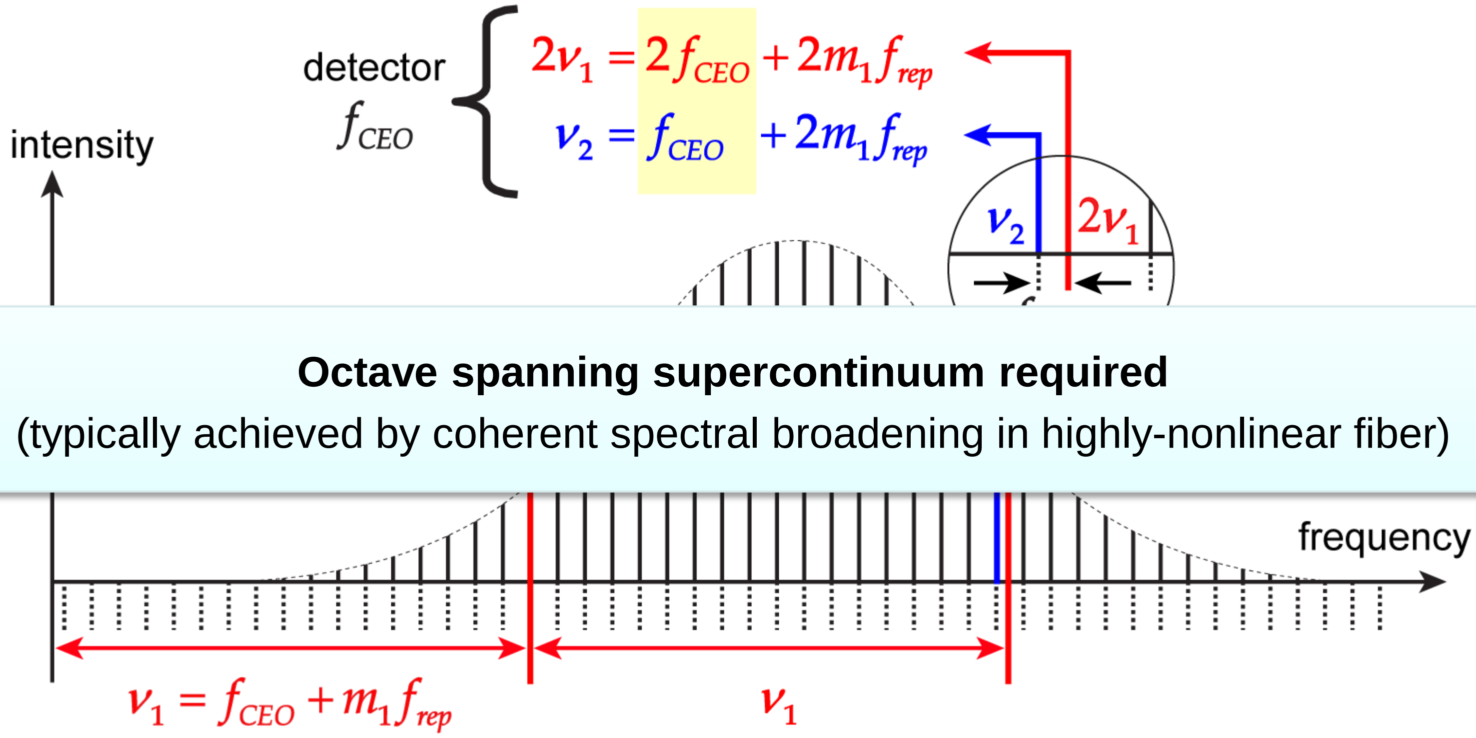
Yb:LuScO TDL: demonstrated first sub-100 fs thin disk laser

C. J. Saraceno, O. H. Heckl, C. R. E. Baer, M. Golling, T. Südmeyer, K. Beil, C. Kränkel, K. Petermann, G. Huber, and U. Keller,"Femtosecond laser oscillators for high-field science", Opt. Exp. 19, (2011)

Detection of the CEO-phase change

- No direct measurement possible
- Indirect f-to-2f-measurement scheme

$$f_{CEO} = \frac{\partial \varphi_{CEO}}{\partial t}$$

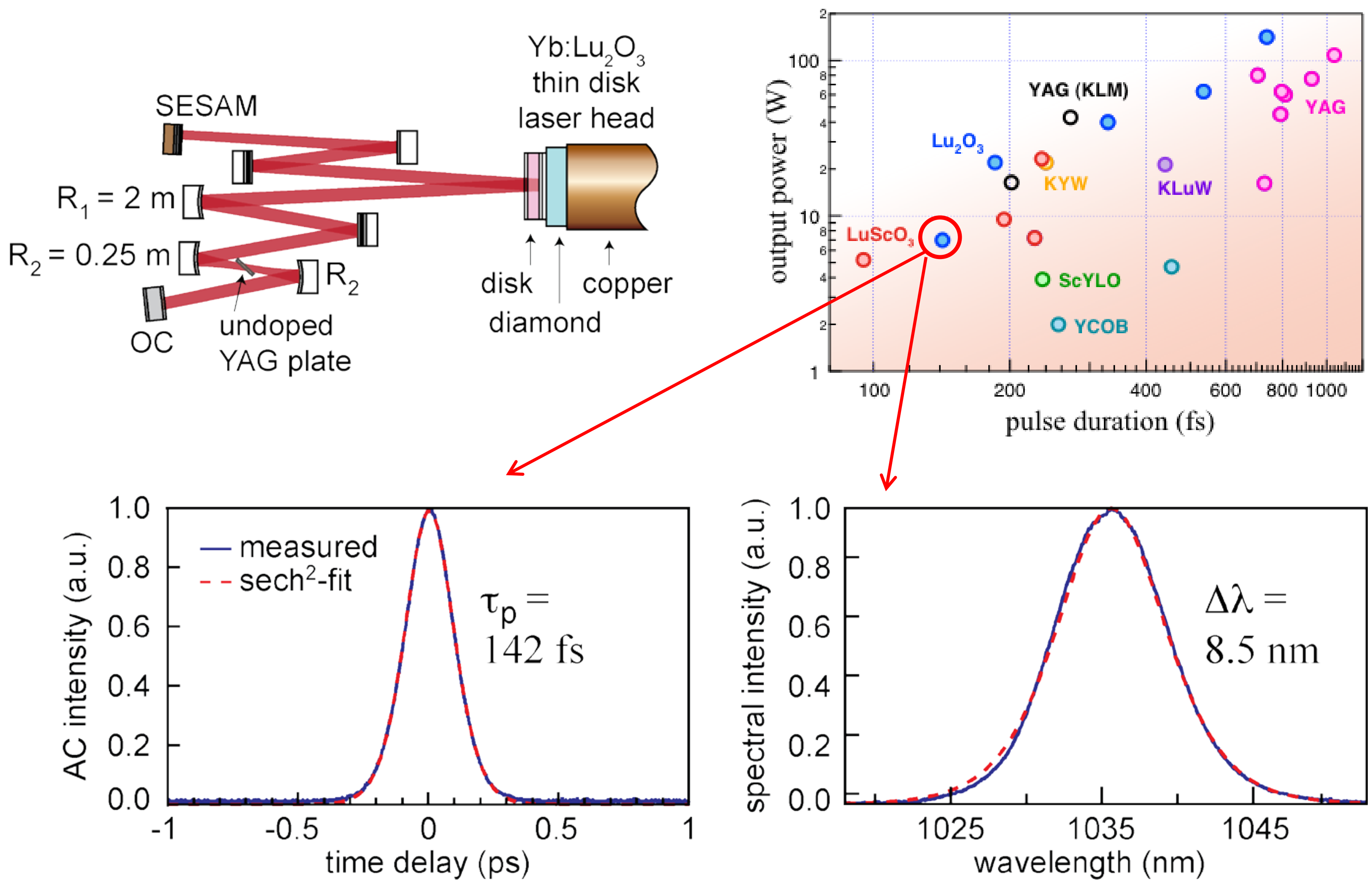


Octave spanning supercontinuum required

(typically achieved by coherent spectral broadening in highly-nonlinear fiber)

H. Telle et al., Appl.Phys. B **69**, 327 (1999)

First CEO detection of a TDL



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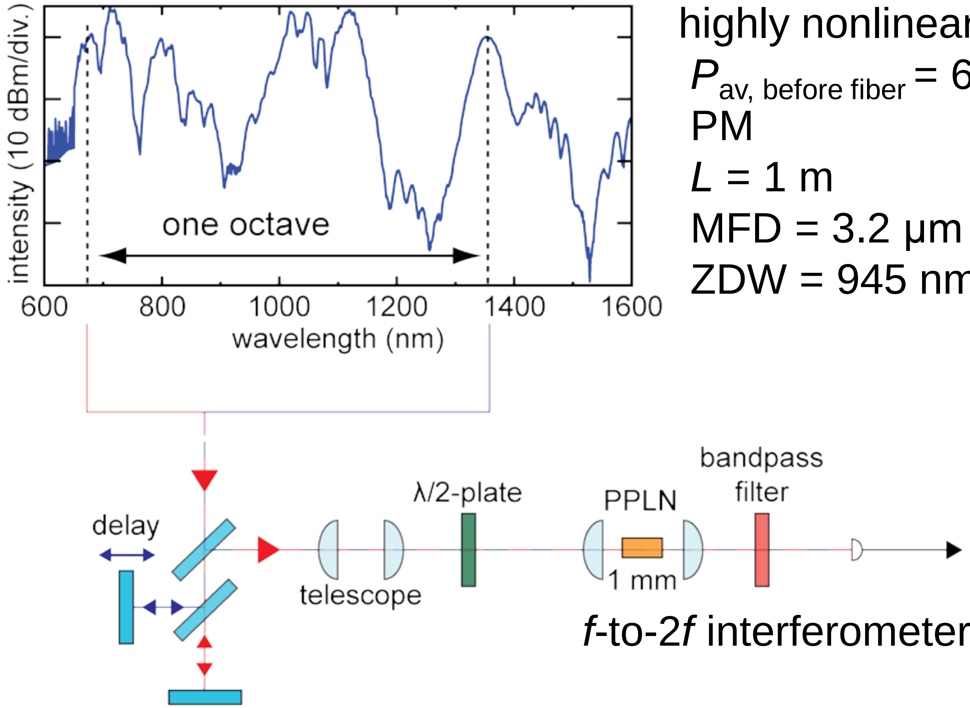
First CEO detection of a TDL



highly nonlinear PCF:

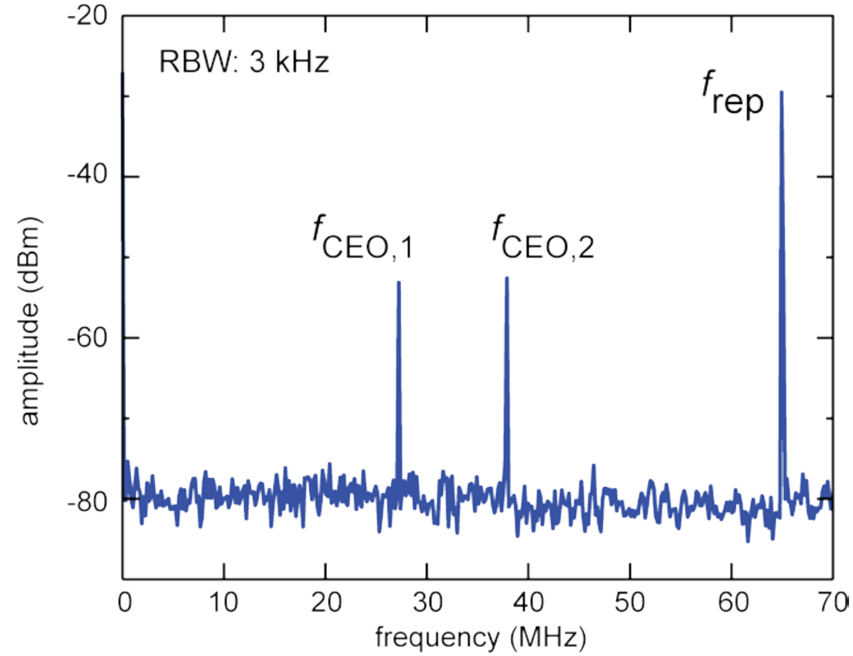
$P_{\text{av, before fiber}}$ = 65 mW

PM

L = 1 m

MFD = 3.2 µm

ZDW = 945 nm



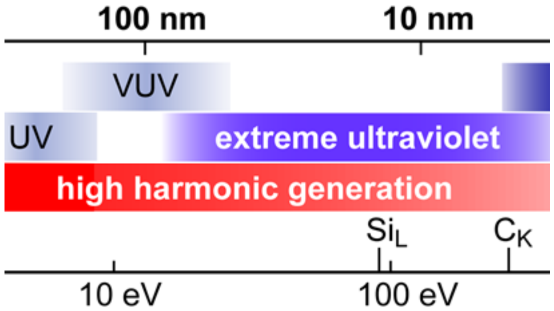
pump current tunable CEO-frequency (slope: 33 kHz/mA) → stabilization

First CEO-beat frequency detection of a thin disk laser (Yb:Lu_2O_3)

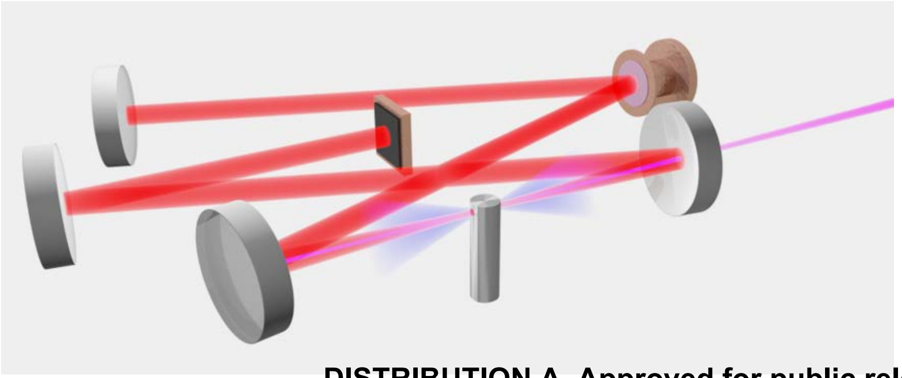
Highest average power self-referenceable frequency comb without amplification

Methodology and impact

- HHG@kHz: huge impact on science
  - as-science
  - imaging semiconductors, biological samples, …
  - dynamics of thermal, acoustic, magnetic properties nanostructures, ...



- Intralaser HHG: novel approach for HHG@MHz
  - Target >20 MHz, >1 mW, fs-duration, 10 nm cut-off
  - high spatial resolution, small focusability
  - resolve fs-dynamics
  - MHz: avoid space-charge effects
  - MHz: frequency comb in VUV/XUV
  - additional IR output for pump probe



- XUV imaging of nanostructures & bio-samples
- frequency comb spectroscopy, e.g. He^+
- ARPES
- …

Appl Phys B (2009) 97: 281–295
DOI 10.1007/s00340-009-3700-z

Applied Physics B
Lasers and Optics

High-power ultrafast thin disk laser oscillators and their potential for sub-100-femtosecond pulse generation

T. Südmeyer · C. Kränkel · C.R.E. Baer · O.H. Heckl · C.J. Saraceno · M. Golling · R. Peters · K. Petermann · G. Huber · U. Keller

Received: 17 April 2009 / Revised version: 26 June 2009 / Published online: 15 September 2009

Abstract Ultrafast thin disk laser oscillators achieve the highest average output powers and pulse energies of any mode-locked laser oscillator technology. The thin disk concept avoids thermal problems occurring in conventional high-power rod or slab lasers and enables high-power TEM_{00} operation with broadband gain materials. Stable and self-starting passive pulse formation is achieved with semiconductor saturable absorber mirrors (SESAMs). The key components of ultrafast thin disk lasers, such as gain material, SESAM, and dispersive cavity mirrors, are all used in reflection. This is an advantage for the generation of ultrashort pulses with excellent temporal, spectral, and spatial properties because the pulses are not affected by large nonlinearities in the oscillator. Output powers close to 100 W and pulse energies above 10 µJ are directly obtained without any additional amplification, which makes these lasers interesting for a growing number of industrial and scientific applications such as material processing or driving experiments in high-field science. Ultrafast thin disk lasers are based on a power-scalable concept, and substantially higher power levels appear feasible. However, both the highest power levels and pulse energies are currently only achieved with Yb:YAG as the gain material, which limits the gain bandwidth and therefore the achievable pulse duration to 700 to 800 fs in efficient thin disk operation. Other Yb-doped gain materials exhibit a larger gain bandwidth and support shorter pulse durations. It is important to evaluate their suitability for power scaling in the thin disk laser geometry. In this paper, we review the development of ultrafast thin disk lasers with shorter pulse durations. We discuss the requirements on the gain materials and compare different Yb-doped host materials. The recently developed sesquioxide materials are particularly promising as they enabled the highest optical-to-optical efficiency (43%) and shortest pulse duration (227 fs) ever achieved with a mode-locked thin disk laser.

PACS 42.55.Rz · 42.55.Xi · 42.60.Fc · 42.65.Re · 42.70.Hj

1 Introduction

Since its first demonstration in the year 2000 [1], the SESAM mode-locked thin disk laser technology achieved higher pulse energies and average power levels than any other mode-locked laser oscillator [2, 3]. The first mode-locked thin disk laser was based on the Yb:YAG gain material and allowed to increase the average power of femtosecond oscillators from previously below 4 to 16 W. The key advantage of the SESAM-mode-locked thin disk laser is its power scalability. The output power can be scaled up by increasing the pump power and mode areas on both gain medium and SESAM by the same factor. Successful power scaling resulted in 80 W average output power [4, 5], which was obtained with an approximately five times larger pump area than in the first mode-locked thin disk laser. The pulse energy of ultrafast thin disk lasers was also successfully increased. Pulse energies as high as 11.3 µJ [6] were generated in a cavity geometry with the thin disk used as simple folding mirror, while up to 25.9 µJ were achieved in an active

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multipass cavity with 13 reflections on the disk [7]. Generating energetic femtosecond pulses directly from a laser oscillator without any further amplification has a number of advantages in terms of simplicity, compactness, and low-noise operation [3]. Based on the previous results, we believe that the ultrafast thin disk laser technology will support further scaling towards several hundred watts of average output power and pulse energies exceeding 100 μJ [3]. So far all these results proving the power scaling possibilities of the thin disk laser concept have been achieved with the gain material Yb:YAG, which is nowadays the best developed thin disk gain material. High-quality disks are available from different manufacturers, because several companies use Yb:YAG thin disks in multikilowatt continuous-wave (CW) laser products. The initial choice of Yb:YAG was not influenced by its suitability to support the generation of femtosecond pulses. It was selected because of its easy growth, high gain cross section compared to other Yb-doped laser materials, relaxed demands on the pump diodes and thermomechanical strength to sustain high power levels. The limited gain bandwidth of Yb:YAG does not support pulse durations shorter than approximately 700 fs in efficient high-power thin disk operation [8] even if pulses as short as 340 fs have been achieved in low-power SESAM-mode-locked laser oscillators [9]. Longer pulse durations can easily be achieved by inserting a spectral filter into the thin disk laser cavity [8]. Using a 60-μm-thick etalon, the pulse duration of an Yb:YAG thin disk laser was continuously tuned over a wide range—from 3.3 to 89 ps—simply by changing the intracavity dispersion [10]. Pulses with a few picoseconds duration are ideally suited for many industrial laser micromachining applications. However, other application fields rely on substantially shorter pulse durations. An important example is that of high-field science experiments, which previously required complex amplifier systems operating at low repetition rates of the order of kilohertz. In contrast, ultrafast thin disk lasers operate at multimegahertz repetition rates and allow for substantially shorter measurement times and an improved signal-to-noise ratio [3]. A promising area for ultrafast thin disk lasers is high-harmonic generation [11, 12] at high power levels. The resulting table-top multimegahertz VUV/XUV sources with high photon flux would have a strong impact in fields as diverse as medicine, biology, chemistry, physics, and material science. Currently, high-field experiments with Yb:YAG thin disk lasers require external pulse compression [13, 14]. Reducing the pulse duration by employing new thin disk gain materials with larger bandwidths is a promising alternative towards lower complexity of the laser system and therefore more compact table-top sources.

Several other laser materials exhibit a larger gain bandwidth than Yb:YAG. Ti:Sapphire material currently holds the record for the shortest generated pulses in mode-locked operation with sub-6-fs pulse duration. Unfortunately, it is not suitable for power scaling to highest power levels because of thermal issues (see Sect. 4). Furthermore, it requires a pump wavelength in the blue-green spectral region, for which no high-power diodes are available. Among the directly diode pumpable gain materials, Yb-doped materials are particularly promising [9, 15, 16]. So far there have been more than ten Yb-doped host materials that generated sub-100-fs pulses in a standard bulk gain medium mode-locked laser oscillator design (see Table 2). Average output power levels of up to 1.5 W were obtained [17]. Unfortunately, it is usually not possible to directly transfer these results to an efficient high-power mode-locked ultrafast thin disk laser. In fact, a suitable gain material for thin disk operation has to fulfill several requirements, not only with respect to its laser properties, but also in terms of thermomechanical strength and other material properties. Furthermore, the technical aspects are challenging: high-quality gain materials have to be manufactured in sufficiently large size, polished to thin disks of 50–300 μm thickness and at least several millimeters diameter, coated, and stress-free mounted onto a heat sink. Due to these challenges only two thin disk gain materials had been mode-locked by the year 2007: Yb:YAG and Yb:KYW [18]. Recently, there has been a large research effort to establish novel thin disk gain materials, which combine the excellent power scaling properties of Yb:YAG with a larger emission bandwidth. Novel thin disk gain materials such as Yb:Lu_2O_3 or Yb:$LuScO_3$ were introduced [19, 20], which resulted in higher optical-to-optical efficiencies and shorter pulse durations than previous ultrafast thin disk lasers [21, 22].

In this paper, we will review gain materials for efficient high-power femtosecond thin disk laser operation. We discuss the demands on these gain materials in terms of laser and thermomechanical properties, and the trade-offs in pulse duration with higher power. We then present an overview of different Yb-doped materials and compare their suitability for the thin disk approach. Finally, we give an outlook towards other new gain materials.

2 Mode-locked thin disk lasers

The development of high-power ultrafast lasers depends strongly on the availability of high-power CW lasers with fundamental transverse mode operation (i.e., TEM_{00} mode). The presence of multiple higher order transverse modes with different frequency spacing leads to mode competition and mode locking instabilities. Achieving fundamental transverse mode operation is particularly challenging for high average powers because of the high thermal load on the intracavity optical components. This is particularly true for the gain medium, in which a substantial amount of heat

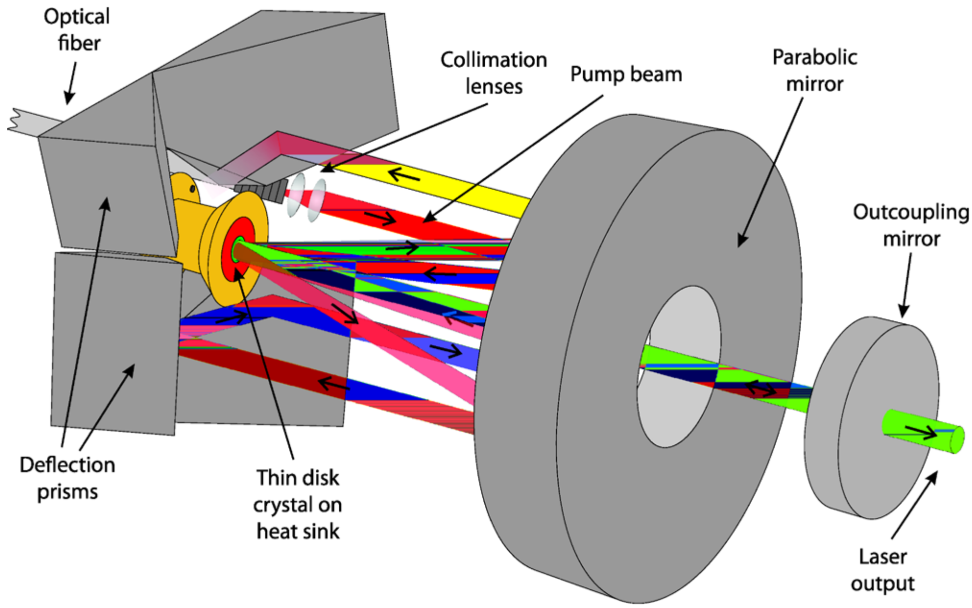

Fig. 1 Schematic setup of the thin disk pump configuration with the disk in the focal plane of a parabolic mirror. The pump optics is usually aligned for 16 or 24 passes through the gain medium

is generated by the pump radiation due to the quantum defect between the pump and laser wavelength and other additional parasitic processes. The resulting thermal gradients can introduce wave front distortions leading to thermal lensing and/or aberrations, which can strongly degrade the transverse beam quality. These problems are usually particularly severe for gain materials with a large emission bandwidth necessary for femtosecond pulse generation. Many of these materials have a disordered crystal structure, and the broad emission spectrum takes its origin in different lattice environments for the laser active ions [23]. However, this disordered lattice restricts the propagation of phonons, which often leads to a significantly lowered thermal conductivity.

During the last decade, two laser geometries have proven to support the highest power levels in fundamental transverse mode operation: thin disk lasers and fiber lasers. Both geometries have excellent heat transport capabilities, because the ratio between cooling surface and active volume is large. Using fiber lasers CW power levels above 1 kW were demonstrated with nearly diffraction-limited transverse mode quality [24]. Nevertheless, they are not ideal candidates for mode-locked femtosecond high-power operation because the large amount of nonlinear effects limits stable pulse formation. Therefore, high-power levels are only achieved with complex amplifier systems [25–27].

The other geometry is the thin disk laser [28], which so far has supported 360 W CW output power in a fundamental transverse mode [29, 30] and 5.3 kW at 65% optical-to-optical efficiency in multimode operation [29]. As the name implies, in this geometry, the gain material is shaped into a very thin disk. Its typical thickness of 50 to 300 µm is small compared to the diameters of the pumped area and the laser beam which are usually a few millimeters. The disk has a highly reflective coating on one side and an antireflective coating on the other side for both, pump and laser wavelength. The reflective side is mounted directly onto a water-cooled heat sink and the disk is used in reflection. The thin disk therefore acts as an active mirror. It can be cooled very efficiently and the heat flow is mainly one-dimensional towards the heat sink, i.e., collinear to the direction of the laser beam. The thermal gradients in the transverse direction are weak, and thermal lensing is therefore strongly reduced compared to a rod or slab design. The pump absorption in a single pass through the thin disk is low but highly efficient operation is achieved by arranging multiple pump passes through the disk (typically 16 or 24 passes), see Fig. 1.

Stable femtosecond pulse formation is achieved using a semiconductor saturable absorber mirror (SESAM [31, 32]), which acts as a nonlinear loss modulator. Such a device exhibits higher reflectivity for higher pulse fluences and therefore favors pulsed over CW operation. The key advantage of the SESAM is its large flexibility to optimize the absorber parameters [33, 34], which prevents mode locking instabilities such as Q-switched mode locking [35]. SESAM designs for high-power thin disk lasers often use a dielectric top coating for the reduction of two-photon absorption (Fig. 2) [6] and an increased damage threshold. The precise measurement of the nonlinear absorption coefficients are discussed in [36, 37].

Ultrafast thin disk lasers usually rely on soliton mode locking [38–40], for which the SESAM only needs to start and stabilize the mode locking process. The final pulse formation is controlled by soliton pulse shaping which balances the negative total intracavity group delay dispersion (GDD) with self-phase modulation (SPM) [15]. Soliton mode locking has the advantage that it relaxes the requirements on the

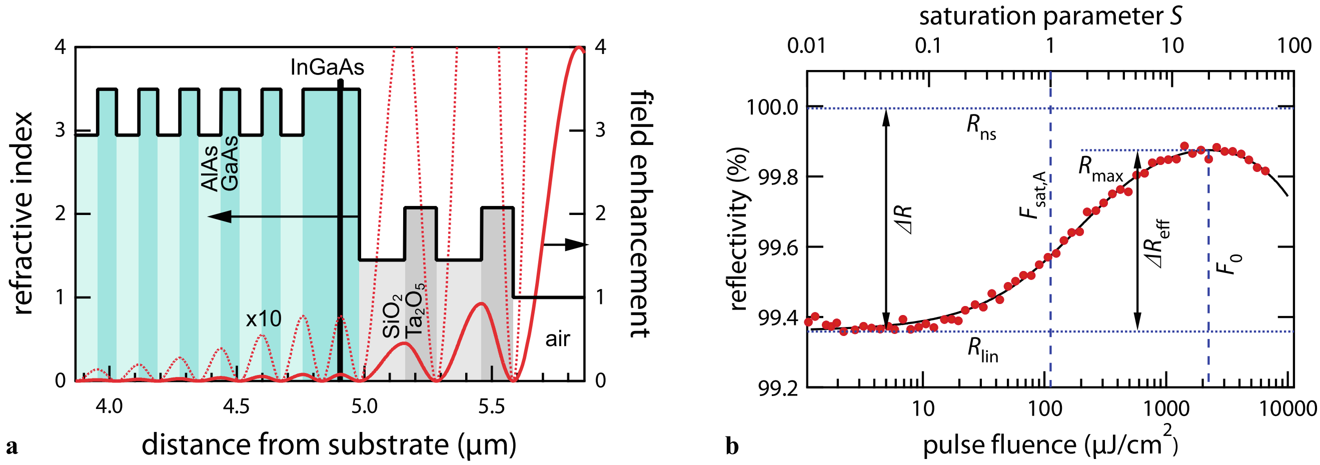

Fig. 2 (**a**) Refractive index pattern and field enhancement of an antiresonant SESAM with a top mirror consisting of two SiO_2/Ta_2O_5-Bragg pairs. The *dotted curve* is the field enhancement multiplied by a factor of ten. (**b**) Reflectivity measurement (dots) as a function of the incident pulse fluence for a SESAM with dielectric top coating. The fit (*solid line*) results in a saturation fluence of $F_{A,sat} = 112\ \mu J/cm^2$, a modulation depth of $\Delta R = 0.64\%$ and non-saturable losses of $\Delta R_{ns} < 0.01\%$. The measurement was done with 570-fs pulses at 1034 nm

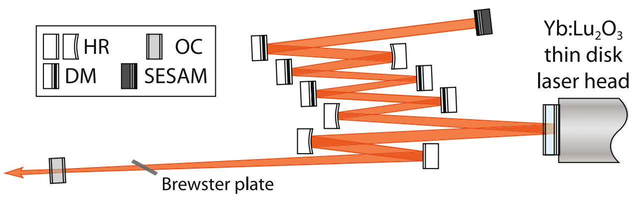

Fig. 3 Schematic of the $Yb:Lu_2O_3$ thin disk laser cavity (not to scale). *HR*: highly reflective mirror, *DM*: dispersive mirror, *OC*: output coupler, *SESAM*: semiconductor saturable absorber mirror

recovery time of the SESAM [41], reduces Q-switching instabilities [35], and generates transform-limited clean soliton pulses even at the highest pulse energies [6]. Chirped-pulse operation in the positive dispersion regime, which is an interesting technique for operating at reduced intracavity intensity levels, has also been investigated with an Yb:KLuW thin disk laser [42]. However, external pulse compression is required and the generated pulses are not ideal transform-limited solitons.

A typical cavity design is shown in Fig. 3 for the $Yb:Lu_2O_3$ thin disk laser discussed in Ref. [21]. The laser setup uses a 250-μm-thick $Yb:Lu_2O_3$ disk, which is used as a folding mirror inside a standing-wave cavity. The disk is pumped by a fiber-coupled diode laser. The pump module is arranged for 24 passes through the gain medium and a pump spot diameter of 1.2 mm. For passive mode locking, a SESAM is used as an end mirror. The nonlinear reflectivity curve of a typical SESAM is shown in Fig. 2. A fused silica Brewster plate is inserted to provide the SPM required for stable soliton pulses and to ensure linear polarization of the laser beam. The outcoupling transmission is 5.2% at the laser wavelength of 1034 nm.

Compared to other solid-state laser designs, the thin disk laser has the least amount of material inside the optical resonator, because all the key components of the passively mode-locked thin disk laser are used in reflection. This is a major advantage for the generation of femtosecond pulses with excellent temporal, spectral, and spatial properties, because the pulses are not affected by excessive Kerr nonlinearity inside the resonator. The concept of the SESAM-mode-locked thin disk laser has the crucial advantage of power scalability. The output power can be scaled up by multiplying the pump power and the mode areas on both gain medium and SESAM by the same factor. The temperature of the thin disk will not rise if the cooling system is capable of removing the additional heat. This scaling procedure avoids damage on the key components because the intensities on the disk and on the absorber remain unchanged. Another important point is that the saturation of the gain and the SESAM remains constant, which is important for preventing mode locking instabilities [8].

3 Disk requirements

It is a challenging task to establish a suitable thin disk gain material for femtosecond pulse generation at several tens of watts of average power. The key requirement is a sufficiently large emission bandwidth, which supports the generation of femtosecond pulses. Another important property is the availability of suitable high-power pump sources. The gain material should exhibit an absorption spectrum accessible to direct pumping by high-power laser diodes. Such diodes are nowadays commercially available at 808, 880, 915, 940,

and 980 nm. The emission bandwidth of the high-power diodes has to match the absorption bandwidth of the material, which might require a stringent selection of the individual diode bars (typically $\Delta\lambda_{\text{diode}} < 3$–5 nm) or additional wavelength stabilization, e.g., with a volume Bragg grating (VBG) (typically $\Delta\lambda_{\text{diode}} < 0.5$ nm) [43, 44]. For optimized thermal management in the laser crystal the gain material should exhibit a small quantum defect, i.e., a laser wavelength close to the pump wavelength. Furthermore, other parasitic heat generating processes should be minimized. Efficient high-power thin disk lasers are usually operated at comparably high inversion levels and it is challenging to suppress parasitic processes, such as nonradiative relaxations, excited state absorption, or up-conversion processes. For low laser threshold and high optical-to-optical efficiency the emission cross section and the fluorescence lifetime have to be large. A high emission cross section is also beneficial to overcome Q-switched mode locking instabilities [35].

It is an advantage of the thin disk laser concept that even materials with a relatively poor thermal conductivity can be used at high power levels [29]. However, to reduce thermal lensing and aberrations, a high thermal conductivity and low temperature dependence of the refractive index is advantageous. Furthermore, the thin disk geometry requires suitable thermomechanical properties of the host material. A good mechanical strength is crucial, not only for laser operation but also for the preparation of thin disks with large diameters. Despite the efficient heat transport in the thin disk approach, strong thermal gradients occur at laser operation, which increase the tendency for thermally induced stress fracture. Furthermore, the absorption of the thin disk has to be large enough to efficiently use the available pump power after the typical 16 or 24 passes through the disk. Besides improving the laser efficiency, a large absorption also helps to prevent damage by the nonabsorbed pump radiation. In the usual pump geometry, the nonabsorbed radiation is reflected backwards and can cause damage of the pump diodes, respectively the fiber output surface of fiber-coupled diodes. If the pump absorption coefficient is low, the required thickness of the disk becomes larger, and the maximum crystal temperature increases. This results in thermal lensing and aberrations, which can prevent TEM_{00} operation and can even lead to damage. Moreover, high temperature compromises the efficiency for three-level laser materials. For a high absorption coefficient a large absorption cross section and a high doping density is required.

In summary, a suitable material should exhibit the following properties:

1. Sufficiently large emission bandwidth $\Delta\lambda_{\text{laser}}$ supporting the generation of femtosecond pulses
2. Pump absorption wavelength λ_{pump} and bandwidth $\Delta\lambda_{\text{pump}}$ matched by available high-power pump lasers
3. Small quantum defect $(1 - \lambda_{\text{pump}}/\lambda_{\text{laser}})$ to minimize heat load
4. Absence of parasitic processes such as nonradiative relaxations, excited state absorption, up-conversion, and cross-relaxation
5. High emission cross section and high fluorescence lifetime to minimize the laser threshold
6. High pump absorption cross sections and high doping concentrations to minimize the disk thickness
7. High thermal conductivity for efficient heat removal
8. Low temperature dependence of the refractive index for low thermal lensing
9. High mechanical strength and weak tendency towards thermally induced stress fracture

4 Gain materials and achieved mode locking results

In Tables 1, 2, we list the properties of a certain number of attractive gain materials with large bandwidths, and some mode locking results that have been achieved. A more detailed list of mode-locked results obtained for various other gain materials is presented in [15]. It is important to emphasize that experiments at low average output power only prove the suitability of a gain material to generate short femtosecond pulses. Unfortunately, one cannot expect to easily transfer such a performance to efficient operation with tens of watts output power. The materials have to fulfill the requirements previously discussed, otherwise the output power will be restricted to a few watts despite the excellent heat transport capabilities of the thin disk geometry.

4.1 Expected pulse duration in the thin disk configuration

Table 2 shows that the pulse durations obtained in low power lasers are usually shorter than in high-power configurations. The minimum achievable pulse duration τ_p for stable soliton mode locking with a SESAM is in detail investigated in [38, 41]. The main limitations are determined by the saturated gain g and the gain bandwidth Δf_g according to

$$\tau_p \propto \frac{g^{3/8}}{(\Delta f_g)^{3/4}}.$$

Compared to low power oscillators, high-power thin disk lasers usually use a large degree of output coupling and operate at a higher saturated gain. Increasing g from 1 to 10% increases the minimum achievable pulse duration by more than a factor of two (if other parameters such as recovery time of the SESAM, its modulation depth, and the maximum acceptable nonlinear phase change of the circulating pulse per roundtrip remain constant, for a detailed discussion see Ref. [41]). Furthermore, the large saturated gain must be achieved from a thin gain medium. This requires

Table 1 Overview of different gain materials for ultrashort pulse generation. The given absorption cross sections for anisotropic materials correspond to the average over all polarizations, which is usually a good approximation in the thin disk pump geometry. To estimate the minimum achievable pulse duration, it is usually not sufficient to consider a FWHM bandwidth of the gain spectrum. Instead, the inversion-dependent shape has to be taken into account (see Fig. 4). Abbreviations: HEM = heat exchanger method, Cz = Czochralski growth, Flux = flux method

Properties	Symbol/Unit	Ti:Sapphire $Ti:Al_2O_3$	Yb:YAG $Yb:Y_3Al_5O_{12}$	Yb:YAB $Yb:YAl_3(BO_3)_4$	Yb:CAlGO $Yb:CaGdAlO_4$
Laser properties					
pump wavelength	λ_P/nm	typ. 488 or 532	940	975	979
absorption bandwidth (FWHM)	$\Delta\lambda_P$/nm	>100 nm	12.5	20	>5
quantum defect	%	67	91	93	96
absorption cross section at λ_P (average)	$\sigma_{abs,P}/(10^{-20}cm^2)$	6	0.83	2.8	1.56
emission cross section at λ_L	$\sigma_{em,L}/(10^{-20}cm^2)$	41	1.89	0.35	0.75
fluorescence lifetime	τ_{rad}/µs	3.2	940	450	420
Other material properties					
thermal conductivity (average)	K/(W/(m K))	33	∼11	4.7	6.6
hardness	Mohs	9	8.5	7.5	–
max. at.% for lasing[a]	at.%	0.25	12	30	13
cation density	$10^{22}/cm^3$	4.7	1.38	0.55	1.25
max. absorption for lasing	α_{abs}/cm^{-1}	7.0	13.7	46.2	25.3
dn/dT	$(dn/dT)/(10^{-6}/K)$	13	9.9	11	–
Symmetry		hexagonal uniaxial	cubic isotropic	trigonal uniaxial	tetragonal uniaxial
Growth		Cz	Cz	Flux	Cz
Reference		[81–83]	[84, 85]	[86, 87]	[88]

Properties	Symbol/Unit	Yb:LuO $Yb:Lu_2O_3$	Yb:ScO $Yb:Sc_2O_3$	Yb:YO $Yb:Y_2O_3$	Yb:LuScO $Yb:LuScO_3$
Laser properties					
pump wavelength	λ_P/nm	976	975	977	975.6
absorption bandwidth (FWHM)	$\Delta\lambda_P$/nm	2.9	2.1	2.1	2.4
quantum defect	%	94	94	95	94
absorption cross section at λ_P (average)	$\sigma_{abs,P}/(10^{-20}cm^2)$	3.06	4.44	2.4	3.3
emission cross section at λ_L	$\sigma_{em,L}/(10^{-20}\ cm^2)$	1.26	1.44	0.92	0.89
fluorescence lifetime	τ_{rad}/µs	870	850	850	850
Other material properties					
thermal conductivity (average)	K/(W/(m K)	12.8	18	13.4	3.9
hardness	Mohs	6–6.5	6.5	6.5	6.5
max. at.% for lasing	at.%	5	5	5	5
cation density	$10^{22}/cm^3$	2.85	3.36	2.69	3.1
max. absorption for lasing	α_{abs}/cm^{-1}	43.6	74.6	32.6	51.2
dn/dT	$(dn/dT)/(10^{-6}/K)$	8.6	9.6	8.5	∼9
Symmetry		cubic isotropic	cubic isotropic	cubic isotropic	cubic isotropic
Growth		HEM	HEM	HEM	HEM
Reference		[67]			

Table 1 (*Continued*)

Properties	Symbol/Unit	Yb:KGW Yb:$KGd(WO_4)_2$	Yb:KYW Yb:$KY(WO_4)_2$	Yb:KLuW Yb:$KLu(WO_4)_2$	Yb:NGW Yb:$NaGd(WO_4)_2$
Laser properties					
pump wavelength	λ_P/nm	981	981	981	975
absorption bandwidth (FWHM)	$\Delta\lambda_P$/nm	3.7	3.5	3.6	8
quantum defect	%	94	94	94	95
absorption cross section at λ_P (average)	$\sigma_{abs,P}/(10^{-20}\ cm^2)$	5.3	6.3	4.4	1.5
emission cross section at λ_L	$\sigma_{em,L}/(10^{-20}\ cm^2)$	2.8 (a)	3 (a)	2.5	~0.1
fluorescence lifetime	τ_{rad}/µs	243	233	254	370
Other material properties					
thermal conductivity (average)	K/(W/(m K))	~3.3	–	~3.3	~1.2
hardness	Mohs	4–5	4–5	–	4.5
max. at.% for lasing	at.%	25	25	25	25
cation density	$10^{22}/cm^3$	0.64	0.63	0.63	0.64
max. absorption for lasing	α_{abs}/cm^{-1}	84.8	94.5	69.3	24
dn/dT	$(dn/dT)/(10^{-6}/K)$	[b]	[b]	[b]	107/42
Symmetry		monoclinic biaxial	monoclinic biaxial	monoclinic biaxial	tetragonal uniaxial
Growth		Flux	Flux	Flux	Cz
Reference		[89]		[72]	[67]

Properties	Symbol/Unit	Yb:YVO Yb:YVO_4	Yb:LuVO Yb:$LuVO_4$	Yb:YCOB Yb:$Ca_3YO(BO_3)_3$	Yb:LSB Yb:$LaSc_3(BO_3)_4$
Laser properties					
pump wavelength	λ_P/nm	985	985	976	981
absorption bandwidth (FWHM)	$\Delta\lambda_P$/nm	7.9	5.8	2.3	>30
quantum defect	%	96	96	94	94
absorption cross section at λ_P (average)	$\sigma_{abs,P}/(10^{-20}\ cm^2)$	3.3	4	1.05	0.51
emission cross section at λ_L	$\sigma_{em,L}/(10^{-20}\ cm^2)$	0.7	0.5	0.45	0.25
fluorescence lifetime	τ_{rad}/µs	310	300	2200	1150
Other material properties					
thermal conductivity (average)	K/(W/(m K))	~7	~8	~2	2.8
hardness	Mohs	5	6	6–6.5	6
max. at.% for lasing	at.%	2	2	35	25
cation density	$10^{22}/cm^3$	1.26	1.31	0.45	0.45
max. absorption for lasing	α_{abs}/cm^{-1}	8.3	10.5	16.5	5.7
dn/dT	(dn/dT) / $(10^{-6}$ /K)	8.5/3.9	–	0.62/0.8/0.17	4.4
Symmetry		tetragonal uniaxial	tetragonal uniaxial	monoclinic biaxial	quasi-trigonal uniaxial
Growth		Cz	Cz	Cz	Cz
Reference			[66]		

[a]Max at.% refers to the maximum doping concentration that can be used for efficient thin disk laser operation. In cases where no max. at.% is given by the crystal structure or in the literature, the value was calculated assuming the same ion density like in Yb:YAG at 12 at.% [90]

[b]Strong variations in the reported values of the thermo-optic coefficients in the potassium tungstates ranging from positive to negative values (depending on the orientation) make it impossible to state reasonable average values

Table 2 Comparison of ultrafast lasers based on different gain materials. A more detailed overview including a discussion of the mode locking techniques is given in Ref. [15]

Laser material		ML technique	λ_0/nm	τ_p/fs	P_{ave}/mW	f_{rep}/MHz	Ref.
Ti:sapphire	$Ti^{3+}:Al_2O_3$	KLM	~800	54	3500	6.23	[91]
		KLM	~800	<40	2500	50	[92]
		KLM	~800	<40	1000	2	[92]
		KLM	~800	<6	120–300	65–85	[47, 48, 93, 94]
Yb:glass	Yb:silicate glass	SESAM	1045	61	65	112	[50]
	Yb:phosphate glass	SESAM	1050	58	65	112	[50]
Yb:YAG	$Yb^{3+}:Y_3Al_5O_{12}$	SESAM	1030	705	80000	57	[4]
		SESAM	1033	340	170	[b]	[9]
		KLM	1051	100	151	150	[95]
		SESAM	1030	2200	8100	63	[96]
Yb:YAB	$Yb:YAl_3(BO_3)_4$	SESAM	1050	87	61	92	[97]
Yb:CALGO	$Yb:CaGdAlO_4$	SESAM	1043	47	38	109	[98]
		SESAM	1043	93	650	27	[99]
Yb:CaF	$Yb:CaF_2$	SESAM	1049	230	1740	[b]	[100]
Yb:LuO	$Yb:Lu_2O_3$	KLM	1032	65	320	99	[64]
		SESAM	1034	535	63000	81	[69, 70]
		SESAM	1035	329	40000	81	[69]
Yb:ScO	$Yb:Sc_2O_3$	SESAM	1044	230	540	86	[101]
Yb:YO	$Yb:Y_2O_3$	KLM	1036	68	540	99	[17]
Yb:LuScO	$Yb:LuScO_3$	SESAM	1036	111	34	92	[102]
		SESAM	1041	227	7200	66.5	[22]
Yb:YO + Yb:ScO		KLM	1041	66	1500	89	[17][a]
Yb:KGW	$Yb^{3+}:KGd(WO_4)_2$	SESAM	1037	100	126	[b]	[103]
		SESAM	1037	176	1100	86	[104]
		SESAM	1039	290	9900	45	[105]
Yb:KYW	$Yb^{3+}:KY(WO_4)_2$	KLM	1046	71	120	110	[106]
		SESAM	1029	240	22000	25	[18]
		SESAM	1043	470	17000	79.8	[107][a]
Yb:KLuW	$Yb:KLu(WO_4)_2$	SESAM	1030	81	70	95	[108]
		SESAM	[b]	490	25600	34.7	[53]
Yb:NGW	$Yb^{3+}:NaGd(WO_4)_2$	SESAM	1032	75	23	[b]	[109]
Yb:NYW	$Yb^{3+}:NaY(WO_4)_2$	SESAM	1035	53	91	96	[110]
Yb:YVO	$Yb:YVO_4$	KLM	1050	61	54	105	[76]
Yb:GdVO	$Yb:GdVO_4$	SESAM	1019	3100	1010	135	[111]
Yb:LuVO	$Yb:LuVO_4$	SESAM	1036	58	85	94	[77]
Yb:YCOB	$Yb:Ca_4YO(BO_3)_3$	SESAM	1056	76	16	94	[112]
Yb:GdCOB	$Yb^{3+}:Ca_4GdO(BO_3)_3$	SESAM	1045	90	40	100	[113]
Yb:LSB	$Yb:LaSc_3(BO_3)_4$	SESAM	1053	58	73	90	[114]
Yb:BOYS	$Yb^{3+}:Sr_3Y(BO_3)_3$	SESAM	1062	69	80	113	[115]
Yb:YSO	$Yb:Y_2SiO_5$	SESAM	1044	198	2600	75	[116]
Yb:SYS	$Yb^{3+}: SrY_4(SiO_4)_3O$	SESAM	1066	70	156	98	[117]
Yb:LSO	$Yb:Lu_2SiO_5$	SESAM	1059	260	2600	75	[116]

[a]Results obtained with two gain media in the resonator. [b]Data not given in the corresponding publication

a high gain per length, i.e., operating the disk at a high inversion level. For many materials, the gain bandwidth Δf_g depends strongly on the inversion level (see Fig. 4), which can additionally increase the minimum achievable pulse duration in the thin disk configuration.

As an example, let us compare the results obtained with Yb:YAG. Its gain bandwidth decreases for high inversion levels: for low level excitation, the gain spectrum is substantially broader with a peak at longer wavelengths (Fig. 4). The low power mode-locked laser in Ref. [9] used a 3.5-mm long 5 at.% Yb-doped YAG crystal. A standing-wave cavity is used, which leads to 7 mm total gain length per cavity roundtrip. Pulses as short as 340 fs were achieved at a center wavelength of 1033 nm. The output power of 110 mW was generated with 2% output coupling. In contrast, the high-power Yb:YAG thin disk laser presented in [4, 5] used an output coupling of 8.5%. It generated 80 W of average power in 705-fs pulses. The laser has a substantially shorter propagation length in the gain medium. The 100-μm disk was 10 at.% Yb-doped and used as a folding mirror, leading to a total gain length of 400 μm per cavity roundtrip. The emission wavelength was 1030 nm, which is 3 nm shorter than for the low power laser. Both effects, the higher saturated gain and the reduced gain bandwidth due to a high inversion level, increased the minimum achievable pulse duration. Please note that for a more precise discussion on the minimum achievable pulse duration, it is important to consider other effects such as spatial hole burning and to investigate the stability limits numerically. A detailed analysis and discussion of pulse formation in ultrafast thin disk lasers is given in Ref. [8].

There are two main directions for achieving shorter pulse duration. Most importantly, the gain bandwidth has to be increased by choosing a better material with larger emission bandwidth. Another possibility is to reduce the saturated gain, which requires operating the laser at low intracavity losses and low output coupling. Especially for higher pulse energies, this may require to eliminate the nonlinearity resulting from the air in the resonator, e.g., by operating the laser in a helium atmosphere or vacuum. In this way, we prevent excessive self-phase modulation, which would otherwise destabilize the pulse formation [45]. The gain per cavity roundtrip can be increased by using a cavity setup with multiple passes through the gain medium. Such resonators were previously used for pulse energy scaling with a resonator in ambient atmosphere because the high saturated gain enables large output coupling, which substantially reduces intracavity SPM [46]. The laser resonator presented in [7] uses 52 passes through a 60-μm thick disk, which leads to a total gain length of 3.1 mm per cavity roundtrip. However, with an output coupling of 78%, it was also operated at high saturated gain and high inversion levels, resulting in 928-fs long pulses. Nevertheless, such multiple-pass cavities are promising for shortening the pulse duration of ultrafast thin disk lasers by enlarging the total gain length per cavity roundtrip. For efficient operation at lower saturated gain, the output coupling and intracavity losses have to be kept sufficiently low. For the above mentioned case, this would require reducing the Kerr nonlinearity of the atmosphere inside the resonator. However, even at very low inversion levels, Yb:YAG does not appear suitable for the generation of sub-100-fs pulses, and alternative gain materials are needed.

4.2 Gain materials with larger emission bandwidth

Ti:sapphire gain material has a particularly high optical gain bandwidth supporting less than 6-fs pulse duration from low energy oscillators [47, 48], which is shorter than for any other laser materials. Unfortunately, its pump wavelength is in the blue-green spectral region, which requires complex and expensive pump lasers. The first CW lasing results with diode pumping were presented very recently, but the obtained output powers were only 8 mW [49]. The development of high-power diode bars operating in the blue-green spectral region is challenging and high-power diode pumping cannot be envisaged in the near future. Furthermore, the material is not well-suited for high-power laser operation at room temperature. The large difference between the emission wavelength and the pump wavelength results in a quantum defect of $>30\%$, which leads to a large amount of deposited heat. The short lifetime results in a high pump threshold, which further increases the heat load in the crystal. So far high doping concentration has not been demonstrated and for the current typical doping concentrations (0.25 at.%) the disk thickness would need to be relatively large even for a 32 pump pass configuration. Therefore, thermal lensing and aberrations would be a challenge even in the thin disk configuration. In summary, Ti:sapphire is not a well-suited material for power scaling in the thin disk approach.

Rare-earth-doped phosphate or silicate glasses also exhibit a very broad emission bandwidth, which supports the generation of pulses with durations of less than 60 fs [50]. Unfortunately, these materials have poor thermal conductivity, which makes them unsuitable for high-power thin disk lasers.

So far, all high-power thin disk lasers used rare-earth-doped crystalline materials such as: garnets (YAG [28], LuAG [51]), sesquioxides (Sc_2O_3 [52], Lu_2O_3 [20], $LuScO_3$ [19]), tungstates (KYW [52], KGW [52], KLuW [53], NGW [54]), vanadates (LuVO [55], YVO [55], GdVO [56]), and borates (YCOB [57], LSB [58]). The doping ions were Yb^{3+} [28], Nd^{3+} [59], Ho^{3+} [60], and Tm^{3+} [61]. In high-power operation, disk diameters of several millimeters up to centimeters are necessary. This requires a growth method that can provide crystals of this diameter in high

optical quality, such as the Czochralski method [62] or the heat exchanger method (HEM) [63]. Ceramic laser materials might be an interesting alternative [64], but at this time superior performance has not yet been proved in the thin disk approach. Furthermore, until recently, only cubic lattices could be produced as ceramics. The first anisotropic ceramics still suffer from low optical quality [65]. Many interesting laser materials grow in an anisotropic crystal structure. This has the advantage of a linearly polarized laser output but at the expense of anisotropy of the thermomechanical properties which increases the tendency for stress fracture in the disk. However, linear polarization in cubic crystals can be easily obtained by several means, for example, by using an intracavity fused quartz plate introduced at Brewster's angle or a thin-film polarizer.

The most successful laser materials for femtosecond pulse generation and direct diode pumping use the rare-earth-ion ytterbium as a dopant. The simple energy level scheme of the Yb^{3+}-ion makes it very attractive for thin disk lasers. The existence of only two Stark manifolds eliminates the occurrence of parasitic processes even at high doping concentrations and the resulting quasi-three-level character leads to a low quantum defect for the laser operation and hence a low thermal load on the active medium. The peak emission cross section is usually in the spectral range of 1030 to 1060 nm, and the zero-phonon-line (ZPL) absorption around 975 nm. The Yb-ion is very suitable for short pulse generation: a relatively strong coupling of the optical $4f$–$4f$-transitions to the phonons of the host leads to broadened absorption and emission spectra which are sufficient for the generation of sub-100-fs pulses in many host materials.

Only in some cases like Yb:YAG, the 940-nm absorption is higher than the ZPL absorption. Today, high-power pump diodes are available for both wavelength ranges. For some materials, the ZPL absorption peak is narrower than the usual emission bandwidth of high-power diodes (3–5 nm). However, during the last years diodes with sufficiently small emission bandwidth have become commercially available. Today power levels of several hundred watts are achieved at <0.5 nm bandwidth from VBG-stabilized laser diodes.

In general, the most suitable hosts for high-power Yb-doped lasers are those where the Yb-ion substitutes Lu. Due to the low mass difference between these ions, the propagation of the phonons, which are mainly responsible for the heat transport in insulators like oxide crystals, is only slightly disturbed. This leads to the lowest decrease in thermal conductivity by the dopant compared to any other ion, including other rare earth sites such as Gd and Y.

The lattice structure also has a strong influence on the thermal material properties. In a disordered lattice different ions are statistically distributed on the same site. This disturbs the propagation of phonons and reduces the thermal conductivity. On the other hand, disordered hosts often exhibit very broad and smooth spectra due to the different lattice environments for the laser active ion.

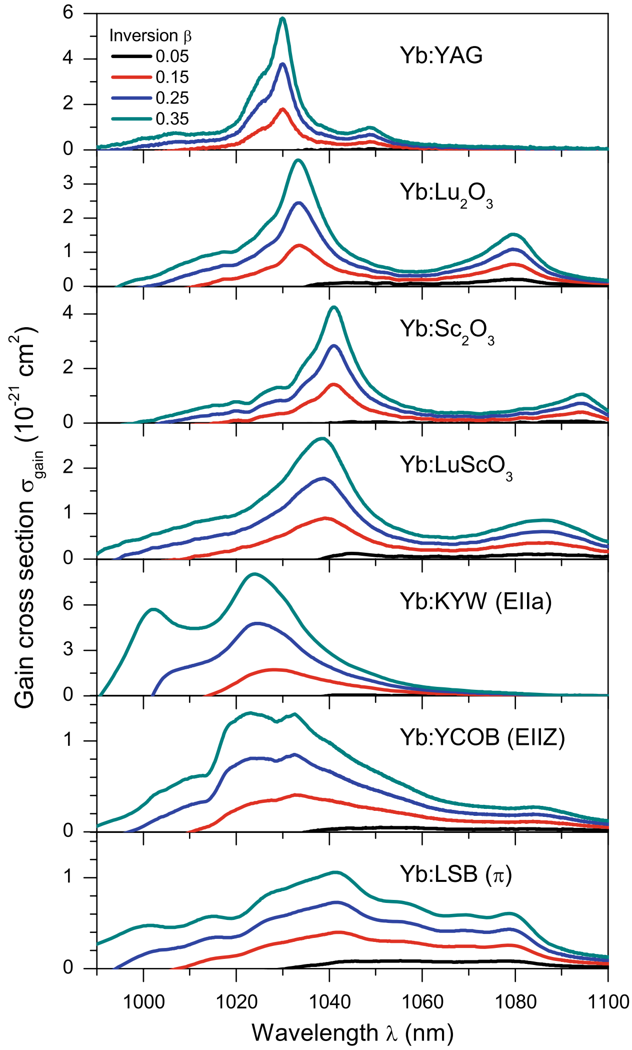

Fig. 4 Gain spectra of the discussed gain materials shown for four different inversion levels β (0.05, 0.15, 0.25, and 0.3, numbers correspond to the total population of the upper laser manifold). For anisotropic materials, the shown spectra correspond to the orientation of maximum gain

5 Yb-doped gain materials for ultrafast thin disk lasers

In the following section, we discuss a number of attractive Yb hosts for ultrafast thin disk lasers. In Fig. 4, we show gain spectra at different inversion levels β (calculated as $\sigma_{\text{gain}} = \beta \cdot \sigma_{\text{em}} - (1-\beta) \cdot \sigma_{\text{abs}}$) for seven Yb-doped materials. The gain spectra were calculated from emission and absorption spectra measured at the ILP Hamburg. Details on these measurements can be found in Refs. [66, 67].

5.1 Yb:garnets

As discussed previously in the text, Yb:YAG is well suited for high-power thin disk lasers, but has a relatively small emission bandwidth among the Yb-doped laser materials. This is a general tendency of all garnet hosts. In the CW-regime Yb:LuAG could be a promising alternative to Yb:YAG because of its better thermal conductivity at high Yb-doping levels. However, it is not promising for generating shorter pulses because its gain bandwidth is even smaller [51].

5.2 Yb:sesquioxides

Another interesting group of gain materials for thin disk lasers are Yb-doped sesquioxides (RE_2O_3 with RE = Y, Sc, or Lu). They were among the first materials to be tested as an alternative to Yb:YAG in the thin disk laser geometry [52]. This is due to their lower quantum defect and good thermo-mechanical properties. They have high thermal conductivity, excellent mechanical stability and a cubic crystal structure. However, for these initial experiments the efficiency proved to be significantly lower compared to Yb:YAG. This was mainly caused by the insufficient crystal quality of the available samples: the high melting point of more than 2400°C is a big challenge for crystal growth. Furthermore, the laser was not pumped at the absorption maximum of 975 nm, but with 940-nm pump diodes.

Recently, the growth process of these materials by the heat exchanger method was optimized, which resulted in Yb-doped sesquioxide crystals of unprecedented optical quality [68]. Furthermore, high-power laser diodes with sufficiently reduced emission bandwidth (<3 nm) became available at 975 nm. This progress made Yb-doped sesquioxides the most efficient thin disk laser materials available nowadays. Optical-to-optical efficiencies well above 70% can be easily achieved in multimode CW operation. Among the different sesquioxides Lu_2O_3 is the most suitable host because it shows the highest thermal conductivity when doped with Yb-ions. For CW operation the output power has recently been scaled up to 149 W [69].

All Yb:sesquioxides exhibit a larger emission bandwidth than Yb:YAG and are therefore more suitable for the generation of ultrashort laser pulses. The first mode-locked Yb:Lu_2O_3 thin disk laser generated 523-fs pulses with an average output power of 24 W [21] at 43% optical-to-optical efficiency, which is higher than for any other mode-locked thin disk laser. Very recently, power-scaling to 63 W in 535-fs-pulses and 40 W in 329-fs pulses was demonstrated [69, 70]. Yb:Sc_2O_3 has not yet been tested in a mode-locked thin disk laser, but we would expect longer pulses than for Yb:Lu_2O_3 due to a slightly narrower emission bandwidth. Yb:Sc_2O_3 has the potential for slightly higher optical-to-optical efficiencies because of its larger emission cross section. However, it might not be as suitable as Lu_2O_3 for further power scaling because of its lower thermal conductivity when doped with Yb [71].

A considerably larger emission bandwidth can be achieved by combining the 7 nm-shifted emission bands of Yb:Lu_2O_3 and Yb:Sc_2O_3 in the mixed sesquioxide Yb:$LuScO_3$ [19]. The resulting material exhibits a more than twice as large emission bandwidth. In initial experiments with this material, pulses as short as 227 fs were obtained, which are the shortest pulses from a thin disk laser [22]. The output power of 7.2 W was limited by growth defects of the only available disk and should be considerably increased with disks of better quality. However, the disordered lattice causes a lower thermal conductivity compared to the pure sesquioxides, which makes it hard to compete with the power scaling potential of Yb:Lu_2O_3, for example.

5.3 Yb:tungstates

The Yb-doped double tungstates $ARE(WO_4)_2$ with an alkali-ion A and a rare-earth ion RE = Gd, Lu, and Y are very interesting hosts for the thin disk laser. The monoclinic tungstates with K as the alkali ion are of particular interest due to their extremely large cross sections [72]. These crystals are among the most efficient Yb-doped laser materials [73]. Another interesting group is represented by the tetragonal tungstates with Na as the alkali ion, as this group crystallizes in a disordered lattice structure, leading to extremely broad spectra. Yb:NYW (Yb:$NaY(WO_4)_2$) and Yb:NGW (Yb:$NGd(WO_4)_2$) are among the Yb-doped materials that delivered the shortest pulses (see Table 2).

A disadvantage of tungstates is that most of them cannot be grown by the Czochralski method due to an incongruent melting behavior. Thus, the flux method has to be used [23], which is a technique with very slow growth rates. Consequently, large crystals of high optical quality are harder to obtain than with the Czochralski or heat exchanger method. Additionally, all of these materials crystallize in a non-cubic structure with a strong anisotropy of the thermomechanical properties, which is disadvantageous for high-power lasers.

One of the few materials of this class that can be grown by the Czochralski method is Yb:NGW (Yb:$NaGd(WO_4)_2$). However, its low thermal conductivity of only 1.1 W/(m K) resulting from the disordered lattice is comparable to that of glasses and not well suited for high-power thin disk lasers. This was experimentally confirmed with a CW multimode thin disk laser for which the output power was restricted to less than 20 W with an optical-to-optical efficiency of 42% [54].

Yb:KYW (Yb:$KY(WO_4)_2$) has been intensively examined for its suitability for the thin disk laser. In CW operation

the output power has been scaled to 73 W at 60% optical-to-optical efficiency [52]. Already in 2002, a thin disk laser based on the Yb:KYW gain material was mode-locked. The laser generated 22 W average output power in 240 fs pulses [18]. Despite these promising initial results, power scaling with this gain material has not yet been demonstrated. Unfortunately, it is difficult to manufacture high-quality thin disks because the host material KYW shows strongly anisotropic thermo-optical and mechanical properties. This challenges stable fundamental mode operation at high pump powers [74, 75].

Yb:KLuW and Yb:KGW exhibit very similar properties compared to those of Yb:KYW. Thin disk laser operation has been demonstrated with both of them [52]. Recently, 440-fs pulses have been realized with an average output power exceeding 25 W with Yb:KLuW. Furthermore, chirped-pulse operation of a thin disk laser oscillator was investigated for the first time with this material [53].

5.4 Yb:vanadates

As can be seen in Table 2, Yb-doped vanadate crystals also enabled the generation of very short femtosecond pulses [76, 77]. Some of these materials such as Yb:LuVO and Yb:YVO have been used in a CW thin disk laser but the output power has not exceeded 15 W [55]. Despite their comparably high thermal conductivity [78] both materials suffer from a very small splitting of the lower Stark level, which makes the laser performance very sensitive to thermal issues. Therefore, these crystals appear not well-suited for the generation of femtosecond pulses in high-power thin disk lasers.

5.5 Yb:borates

Many Yb-doped borates exhibit a disordered crystal structure that can lead to broad spectra. Among the different borates, Yb:YCOB and Yb:LSB have the advantage of a nearly congruent melting behavior, which enables Czochralski growth [79, 80]. Both materials support the generation of sub-100-fs pulses in the low power SESAM-mode-locked regime (see Table 2). They have quite low absorption coefficients compared to the other mentioned thin disk laser materials. Therefore, rather thick disks exceeding 0.5 mm were used in CW thin disk lasers. Despite this fact, output powers exceeding 25 W were realized without any thermal roll-over (LSB: 40 W, YCOB: 26 W) [57, 58]. So far, these promising materials have not been used in ultrafast thin disk lasers, but mode-locked operation with sub-200-fs pulses and several tens of watts average power appears feasible.

6 Conclusion

Ultrafast SESAM-mode-locked thin disk lasers offer a robust and power-scalable solution to the challenges of generating femtosecond pulses at high power levels without external amplification. The efficient heat removal in the thin disk laser geometry minimizes thermal lensing and aberrations. This enables high power levels in a fundamental transverse mode even for gain materials with relatively low thermal conductivity as is often the case for disordered gain materials with a broad emission bandwidth. Due to its design flexibility, the SESAM is an ideal device for mode locking at such high power levels and pulse energies. The concept of the SESAM-mode-locked thin disk laser has the essential advantage of power scalability: the output power can be scaled up by increasing pump power and mode areas on both gain medium and SESAM by the same factor. The properties of the employed gain material play a crucial role because they determine both the minimum obtainable pulse duration and the achievable output power levels.

Today the highest output powers and pulse energies are still obtained with the garnet Yb:YAG, which was the first thin disk gain material. But with the advances in research and development of new Yb-doped hosts and the availability of suitable pump diodes operating in the 980-nm spectral region, other gain materials have the potential to outperform Yb:YAG in the area of ultrafast pulse generation. However, finding a suitable gain material is not trivial: an ideal femtosecond thin disk gain material has to excel not only in terms of laser properties but also in terms of other material characteristics. In this paper, we discussed gain material requirements and reviewed the most promising candidates. Yb^{3+}-doped gain materials have proven to be well suited for thin disk lasers thanks to their low quantum defect and broad amplification bandwidths allowing femtosecond pulse generation. Many crystalline oxide host materials provide thermomechanical properties that are well suited for high-power laser systems.

We expect that femtosecond pulses with several hundred watts of average power will be achieved in the near future. Besides the well-established Yb:YAG gain medium, the sesquioxide Yb:Lu_2O_3 is a promising material for this task. It has already proven its high suitability for the thin disk approach achieving the highest optical-to-optical efficiency of any thin disk laser both in CW multimode (i.e., 73%) and mode-locked operation (i.e., 43%).

Currently, the shortest pulses obtained from an ultrafast thin disk laser are 227 fs achieved with Yb:$LuScO_3$, another sesquioxide. Other Yb hosts with broader emission spectra appear even more promising to further reduce the pulse duration. Borates such as Yb:LSB and Yb:YCOB are particularly promising. Operation at several tens of watts in the CW regime has been demonstrated, but no mode locking has been demonstrated so far in a thin disk laser. Recently, other established femtosecond laser materials such as Yb:CALGO or different Yb:silicates (see Table 2) also appear promising to extend the performance of ultrafast thin disk lasers into the sub-100-fs regime.

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